The Radioactivity of Naturally Occurring Isotopes with $Z < 82$.

by D.E. WATT.

This thesis is being submitted to the University of Glasgow for the degree of Doctor of Philosophy.

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PREFACE.

1. General.

This thesis describes the techniques used and the results obtained in a search for the long-lived naturally occurring radioactive isotopes with atomic number <82 and excludes the uranium, thorium and actinium series.

The work was started in October 1953 and largely completed by 1956. However as the thesis is only now being submitted (1961) an effort has been made to bring the contents up to date wherever applicable. Thus chapter two presents the current ideas on the reduction of counter backgrounds. Recent work by the author on V-50 (Paper 2 Chapter 3) has been introduced as this completely alters the indications of the earlier results reported in 1956. Results of a recent measurement for the β-ray maximum energies of In-115 and Re-187 are included to enable a complete statement on the double isobars (Chapter 4) to be made. Full references until June 1961 are given where relevent.

The discovery of parity non-conservation by Lee and Yang in 1956 does not affect the present application of β-decay theory and is not considered except in Chapter 6 where it has an important contribution to the discussion on double beta decay.

2. Division of Work.

The author (D.E.W.) is solely responsible for all aspects of the work comprising Chapter 2, (except in the background tests sect^{n} .2.5 where Ramsden cooperated) and the alpha activity measurements in Chapter 5. In Chapter 3 on the triple isobars, the experiments on La-138, V-50 (Paper 1) and Lu-176 were carried out in collaboration with R.N. Glover (R.N.G.) who wrote the final papers. Interpretation and experimentation on these isobars was shared equally. D.E.W. was responsible for the design, interpretation and reporting of the "Half-Life of V-50" where R.L.G. Keith assisted on the experimental side and with the calculations.

In Chapter 4, on the double isobars, R.N.G. and D.E.W. shared in the half-life measurements on Cd-113, Sb-123, In-115 and Te-123. D.E.W. was responsible for the later work on the maximum β-energies of In-115 and Re-187 and the measurements of the γ-ray half-life limits for Sb-123 and Te-123. Interpretation and reporting of the results was by D.E.W.

3. Published Papers.

Some of the author's work described in this thesis has been accepted for publication in the journals. These papers are listed below.

PREFACE (Continued).

1. The Natural Radioactivity of Lanthanum Phil Mag 2 49 1957.

2. A Search for Natural Radioactivity in Vanadium Phil Mag 2 May 1957.

3. A Search for Electron Capture in Lu-176 Phil Mag 2 May 1957.

4. The Half-Life of V-50 - to be published in Nuclear Physics.

5. A Search for Radioactivity among the Naturally Occuring Pairs Cd-In-113, In-Sn-115, Sb-Te-123 and Re-Os-187. to be published in Philosophical Magazine.

6. A Low Background Laboratory. Int. Journ. of Appl. Radn and Isotopes 1961.

July, 1961.

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CONTENTS.

ABSTRACT.

Consideration of nuclear energies and stability characteristics lead to the conclusion that in cases where three neighbouring isobars, or a double isobar, occur in nature then the odd-odd member of the triple isobar and one member of the double isobar must be radioactive. Since these nuclides are expected to have very long half-lives, greater than the age of the earth, sensitive detection techniques, capable of measuring low specifice activities are necessary. Some aspects of these are discussed.

The nature of the cosmic radiation is considered in a fundamental approach to the form of shielding required to produce the optimum in low background counting. Contributing factors, of a secondary nature, to the background counting rate in proportional and scintillation counters are discussed and suggestions given on how these can be removed. A background of 0.33 counts/min/litre above 500 eV at one atmosphere pressure was obtained for a wall-less proportional counter with an arrangement using boron-loaded paraffin wax inside a low background laboratory It is concluded that this residual count rate is due to -radiation which would be greatly reduced with a more efficient -ray detector in the anti-coincidence system.

The characteristics of an underground laboratory and of a specially built low-background cell are described.

Proportional and scintillation counting techniques are used in an investigation of, and search for, natural radioactive species below atomic number 82.

ABSTRACT (Continued).

The existence of two γ-rays at 0.81 ± 0.01 Mev and 1.44 ± 0.02 Mev in La-138 is confirmed. No annihilation quanta were observed. Coincidence studies show that these γ-rays are not in cascade but that the 1.44 Mev transition is in coincidence with the electron capture branch. A previously unobserved β-ray with maximum energy of 205 ± 10 Kev was detected. The partial half-life of the electron capture branch is found to be $(1.64 \pm 0.06) \times 10^{11}$ years. From the β-ray specific activity the half-life is $(4.1 \pm 0.7) \times 10^{11}$ years and from the 0.81 Mev γ-ray is $(3.5 \pm 0.3) \times 10^{11}$ years. A log ft value of 19 is calculated for the β branch indicating the transition is 3rd forbidden.

Two experiments were carried out on V-50. In the first a half-life of $(4.0 \pm 1.1) \times 10^{14}$ years was obtained using a two inch NaI crystal to detect the γ-rays from a sample known to contain the normally experienced amount of Uranium-Thorium-Actinium series, as impurity. Later a re-investigation of this isotope using a combination of a 9 inch x 6 inch NaI crystal with five kilograms of spectrographically pure materials, which gave a factor of twenty increase in sensitivity over the best of the previous methods, proved the half-life to be $1.8 \pm 0.4 \times 10^{16}$ years for negatron emission and $9.6 \pm 1.6 \times 10^{15}$ years for electron capture. It is shown that the degree to which natural uranium and thorium occur in the source material must be accounted for in the measurement of very low specific activities,

ABSTRACT (Continued).

The importance of correcting for changes in the natural background spectrum shape caused by the different absorption for the cosmic radiation and local radioactivity in the presence of large amounts of source material are emphasised.

A search was undertaken for γ-rays or conversion electrons which may be associated with the expected, but so far unobserved, electron capture branch of Lu-176. It is concluded that the upper limit for this mode of decay is $(3 \pm 1)\%$. The half-life of the β branch obtained via measurements with the associated γ-rays is $(2.1 \pm 0.2) \times 10^{10}$ years.

Among the double isobars investigations on Cd-113 and Sb-123 show these to have minimum βhalf-lives of 1.3×10^{15} years and 1.32×10^{16} years respectively. A new value of 480 ± 30 Kev has been obtained for the beta disintegration energy of In-115 and the half-life confirmed to be $6.0 \pm 0.4 \times 10^{14}$ years. The K electron capture of Te-123 has been observed for the first time. A half-life of $1.24 \pm 0.10 \times 10^{13}$ years is calculated. The beta decay of Re-187 has been observed to have a maximum β energy of 1.2 ± 0.10 Kev and an estimated half-life of 3×10^{10} years. This is the sofest β-ray spectrum ever to be detected. Lower limits for gamma ray emission from In-113, Sb-123 and Te-123 are respectively 1.4×10^{15} years, 1.5×10^{15} years and 2.5×10^{13} years.

The systematics of natural α-emitters among the medium heavy elements are discussed and a proportional counter metho

ABSTRACT (Continued).

/method with the potential of measuring α half-lives up to 10^{20} years is described. Limitations on this method are the levels of natural uranium, thorium and actinium impurities, in most substances.

Finally, suggestions are given for some future experiements which should clarify the position regarding the natural radioactive substances below lead. The significance and possible detection of Double Beta Decay is considered.

Chapter 1.

INTRODUCTION.

1.1 The Stability of Nuclei.

If a chart of the nuclides is constructed by plotting the atomic number Z, as the ordinate and the neutron number, N, as the abscissa it is apparent that the stable nuclides below A = 36 lie along the line N= Z. For higher mass numbers the stable nuclei have a progressively increasing neutron excess. The explanation for this deviation from the N=Z line lies in the fact that as A increases the number of protons (Z) in the nucleus increases and there is a corresponding rise in the electrostatic repulsive forces until above Z = 82 instability results. Excess neutrons are required to counteract the coulomb forces.

It is found[1] that among the light nuclides (Z<20) those with an equal number of protons and neutrons are exceptionally stable e.g. He_2^4, C_6^{12}, and O_8^{16}. A plot of binding energy per particle against mass number shows that much larger energies are required to extract a particle from these nuclei than for higher Z isotopes. Peaks which occur in the curve are caused by the completion of nuclear shells at the 'magic numbers' of neutrons or protons. Above Z=20 the coulomb forces become significant and account for the gradual average decrease in binding energy per particle as Z increases.

Unstable nuclides lying above the 'line of stability' decay by β^+ emission or by electron capture whilst those below the curve transform by β^--decay for constant A values i.e. the nuclide

/nuclides tend toward greater stability.

An alternative representation[2] of the stability characteristics of nuclei is given by a three-dimensional plot of Z, N and the true mass, M, of the nucleus. For odd A nuclei the section of the valley with the unstable nuclei on the slopes. A cross-section of the valley for nuclides with constant A has a parabolic shape with the minimum position (See fig. 1) corresponding to some value of nuclear charge Z_o which determines the most stable position. Since this value is unlikely to be integral that value of Z which lies nearest to Z_o determines the stable isobar of odd A. In all cases, with the exception of Cd-113; In-113; Sb-123 Te-123; Re-187 Os-187, there is only one stable isobar for each odd A. In the case of the exceptions it is believed that one of each pair is unstable. Te-123 and Re-187 are shown in this work to decay by electron capture and -respectively. Also In-115 and Rb-87 are part of naturally occurring pairs of odd A isotopes and have been verified as unstable.

For even A nuclei, the graphical representation reveals the existence of two parabolas - one for odd-odd nuclei and one for even-even nuclei. β -decay energetics have proved these to be separated in energy by about $\frac{68}{A^{\frac{3}{4}}}$ Mev and to span the odd A curve which comes in the middle. Since the curve of even-even isotope is at a lower mass or energy level they are more stable and the odd-odd nuclides can be expected to be less stable. The explanation can be found in the Pauli exclusion principle whereby two like particles (e.g. either protons or neutrons) may/

/may occupy the same nuclear energy state provided the wave functions are anti-symmetric. It follows that isobars with even numbers of protons and neutrons will be very stable which accounts for the stability of He_2^4, C_6^{12}, and O_8^{16}. Also because of the saturation characteristic of nuclear forces each nucleon interacts strongly within the state and weakly outside it thus explaining the increased stability of nuclei with closed shells.

If two neighbouring insobars have not identical masses and differ in charge by only one unit then one or other must be radioactive provided that $M_Z - M_{Z+1}$ is greater than the energy required to remove a valency electron from the atom with charge Z+1 in which case β-decay is possible. Alternatively if $M_{Z+1} - M_Z$ is greater than the K shell ionisation potential of the atom M_Z, K electron-capture can occur.

Similarly if two stable isobars differ in charge by two units then the odd-odd members of the naturally occurring triple isobars e.g. K-40, V-50, La-138, Lu-176, and Ta-180, are expected to decay by β- and K capture respectively. All of these with the exception of Ta-180 have been proved active. It is significant that many of these naturally occurring double and triple isobars are close to closed neutron or proton shells.

The foregoing discussion is consistent with the frequency of occurrence of the various stable isotopes. For example, there are three times as many even A nuclides as there are odd A nuclides. The number of nuclei with even Z and odd N is about equal to those with odd Z and even N. All nuclei with/

/with even A have even Z except the very light isotopes H_1^2, Li_3^6, B_5^{10} and N_7^{14} whilst elements with even Z all have many species with even A but only a few with odd A. No element except tin, on the closed proton shell at Z=50, has more than two isotopes of odd mass number, Odd Z elements never have more than two stable isotopes and these are all odd mass numbers above N_7^{14} with the exception of Ta-180 which has not yet been proved to be radioactive. Above Z=7, there are no odd-odd stable nuclei in nature (once again with the exception of Ta-180).

Statistics[3] on the relative abundances of the isotopes is informative regarding the stability of the elements. Mayer finds that the isotopic abundances are not distributed about the centre for specific elements and that the lowest abundances occur on the neutron deficient side. The concentrations for the lightest of these is <2% although there are exceptions to the rule in the region of closed shells at N=50 and 82. Also, elements with even Z>40 have no abundances >35% except Sr-88, Ba-138 and Ce-140, each with closed neutron shells.

The average number of stable isotopes for odd neutron numbers is <1 but for N=50 there are six and for N=82 there are seven. The stable calcium isotopes with Z=20 have a much larger than usual mass range of 8 neutrons and tin at Z=50 has ten isotopes with a range of 12 neutrons.

To summarise, from qualitative reasoning it is deduced that one member of the isobaric pairs Rb-Sr-87, Cd-In-113, In-Sn-115, Sb-Te-123 and Re-Os-187, should be unstable with

/with respect to β- or electron-capture transitions and the odd-odd member of each set of triple isobars viz K-40, V-50, La-138, Lu-176, Ta-180 should decay to their respective even-even neighbours.

These conclusions can be deduced from the semi-empirical mass formula described in the following section.

1.2 The Semi-Empirical Formula.

The parabolic-shaped curves, obtained by plotting either binding energy against the neutron excess $\frac{1}{2}(N-Z)$, or total mass with charge number, for any set of isobars is reproduced in several of the semi-empirical mass formulae, one of which has been derived by Weizsacker.

It is assumed that the binding-energy of the nucleus is caused by forces, analogous to those exerted on a liquid drop. Consequently there is a volume-interaction term = $a_1 A$, caused by the internally saturated forces acting among neighbouring nuclei, which is diminished to some extent by the unsaturated forces at the surface. This latter force is a form of surface tension and is represented by a term proportional to the square of the nuclear radius, i.e. = $a_2 A^{2/3}$.

The existence of the electric charge associated with the protons in the nucleus creates coulomb repulsive forces which further diminish the binding energy. This charge interaction term = $a_2 Z (Z-1) A^{-1/3}$, where the $A^{-1/3}$ factor expresses the dependence of the energy contribution on the nucleur radius.

In addition to the forces analogous to those experienced by a charged liquid drop there are two further terms caused by purely nuclear effects. These are

1) A term to allow for the effects of symmetry properties of the nuclear states and how these are modified when the number of nucleons is kept constant and the neutron excess is varied. This is expressed as $a_4\frac{\{A/2-Z\}^2}{A}$ and

2) A pairing term to allow for the fact that nucleons show a strong interaction in pairs for any given kind although a proton and neutron do not interact appreciably in different quantum states. For even-even nuclei the term is $+ \delta/A^{3/4}$, for odd-odd = $- \delta/A^{3/4}$ and for odd-even, or even-odd is zero.

Hence the binding energy of a nucleus can be expressed as

$$B(Z,N) = a_1 A - a_2 A^{2/3} - a_3 Z(Z-1)A^{-1/3} - a_4 \frac{(A/2 - Z)^2}{A}$$

$$\begin{cases} + \delta A^{-3/4} & \text{even-even} \\ & \text{even-odd} \\ \pm 0 \quad \text{or} & \text{odd-even} \\ - \delta A^{-3/4} & \text{odd-odd} \end{cases}$$

A best fit to the experimental data is given by

$a_1 = 14.0$ Mev, $a_2 = 14.0$ Mev, $a_3 = 0.61$ Mev, $a_4 = 84.2$ Mev and $\delta \sim 34$ Mev.

This equation, apart from the spin term, is a quadratic in Z for a given A and hence a plot of B(Z,N) against Z for a series of isobars is a parabola.

Coryell[4] expresses the pairing energy slightly differently to help in distinguishing between even Z, odd N and odd Z, even N nuclei.

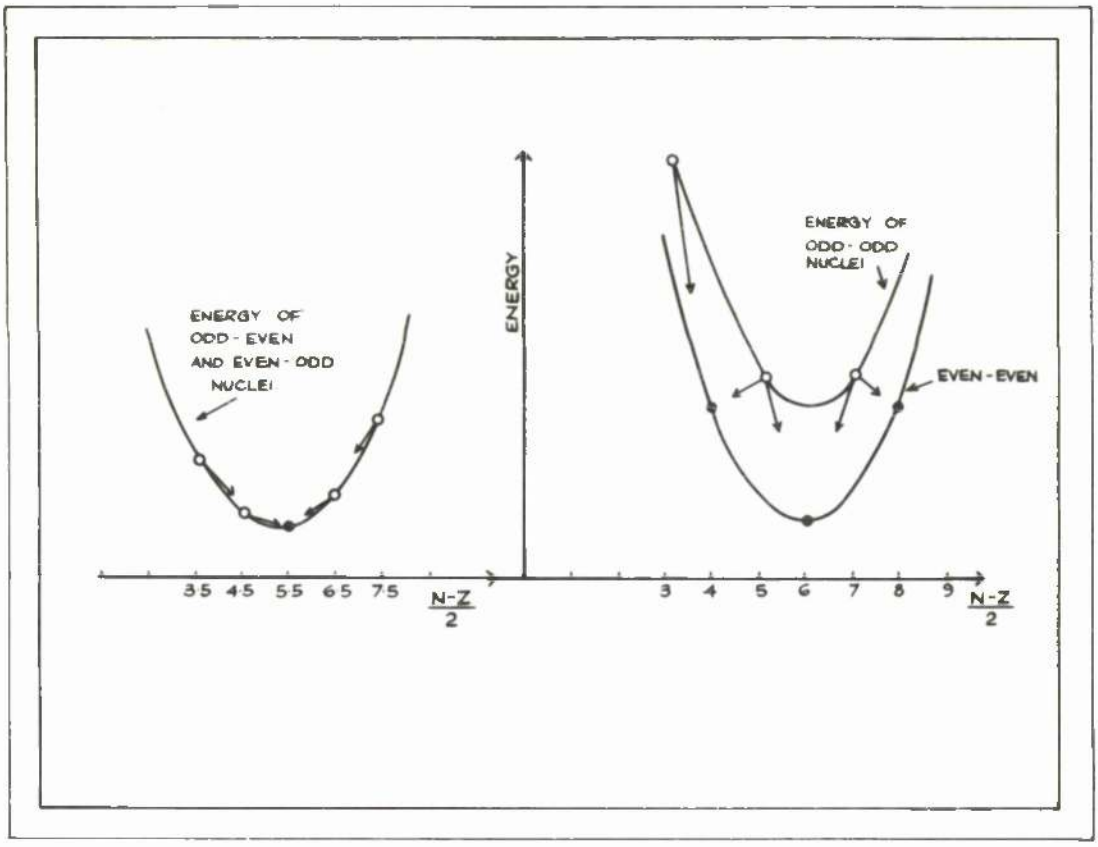
ENERGY OF
ODD - EVEN
AND EVEN - ODD
NUCLEI
ENERGY
ENERGY OF
ODD - ODD
NUCLEI
EVEN - EVEN
35 45 55 65 75
N-Z
2
3 4 5 6 7 8 9
N-Z
2

FIG 1.

If the pairing energy for two protons is 2π, the unpaired proton of an odd Z nucleus will introduce an instability term of π compared to the paired terms and similarly the odd neutron in an odd N nucleus contributes ν to instability. Hence the pairing or spin terms are represented by

$$\begin{cases} \pi+\nu \sim 2\delta & \text{A even, N, Z are odd.} \\ \pi \sim \delta & \text{Aodd, Z odd and N even.} \\ \nu \sim \delta & \text{A odd, Z even and N odd.} \\ 0 & \text{A,N,Z are all even.} \end{cases}$$

If B_o and Z_o are the minimum values of the binding energy and the charge (non-integer in this case) on the parabola then the total energy for β-emission is given by

$$Q_\beta = B_o\,(Z_o - Z - 0.51) \mp \delta_A \qquad \text{(A even; even Z takes the negative sign)}$$

where $\delta_A \sim \pi+\nu$ is the difference between the two surfaces (fig.1) similarly for odd A species

$$Q_\beta = B_o\,(Z_o - Z - 0.51) \mp (\pi-\nu)$$

It is apparent that the condition for stability of an odd A nuclide of even charge Z is satisfied if $-0.51 - \frac{\pi-\nu}{B_o} < Z_A - Z < 0.51 + \frac{\pi-\nu}{B_o}$. Hence if we assume the mass of the neutrino is zero the occurrence of two stable isobars of odd A differing in charge by one unit is impossible.

Similar arguments apply to proton and electron-capture decay.

In the case of even A nuclides, the requirement for stability is that if $\delta_A/B_o > /Z_o - Z/ - 0.5$ then the nucleus is stable and if $\delta_A/B_o < /Z_o - Z/ - 0.5$ then the nucleus in unstable.

The semi-empirical mass formula permits a rough calculation of nuclear masses for a large number of nuclei. However it deviates markedly from experimentally measured atomic masses in the region of closed shells and is unsatisfactory formany calculations.

Harris B. Levy[5] has developed a new empirical mass equation in which he treats each shell region individually and takes into consideration all the linear relationships between Z_A and A, E and A for constant I, neutron binding energy and A for constant Z, the parabolic relationship M and Z for constant A, and the mass-defect M-A and A constant Z. This formula is simply expressed as

$$M(U,V) = C_0 + C_1U + C_2V + C_3\,UV + C_4U^2 + C_5V^2 \text{ ------ (2)}$$

where U and V are any two of the quantities Z, N, A and I. The mass defect is expressed in terms of A and Z as

$$\Delta M(A,Z) = \alpha_0 + \alpha_1 A + \alpha_2 Z + \alpha_3 AZ + \alpha_4 Z^2 + \alpha_5 A^2 + \text{f} \text{ ---- (3)}$$

depends on whether the nuclide is odd-odd, even-even, even-odd or odd-even and the coefficients are chosen to give a best fit with experiment, there being a separate set for each shell region. The calculated results are reasonably accurate since 84% agree to within 0.25 Mev and 95% to within 500 Kev. In cases where double or triple isobars exist, Equation (3) is sometimes useful in predicting which nuclide will be the active one.

1.3 Naturally Occurring Isobaric Pairs and Triplets.

The preceding discussion on nuclear systematics has indicated that in cases where isobaric doublets or triplets exist in nature, then one isobar in each group must be radioactive. In Table I are listed all such groups with Z<82. (The well-known natural radioactive series from U^{235}, U^{238} and Th^{232} are omitted from this discussion).

Table I.

Triple Isobars

$A_{18}^{40} - K_{19}^{40} - Ca_{20}^{40}$

$Ti_{22}^{50} - V_{23}^{50} - Cr_{24}^{50}$

$Ba_{56}^{138} - La_{57}^{138} - Ce_{58}^{138}$

$Yb_{70}^{176} - Lu_{71}^{176} - Hf_{72}^{176}$

$Hf_{72}^{180} - Ta_{73}^{180} - W_{74}^{180}$

$Rb_{37}^{87} - Sr_{38}^{87}$

$Cd_{48}^{113} - In_{49}^{113}$

$In_{49}^{115} - Sn_{50}^{115}$

$Sb_{51}^{123} - Te_{52}^{123}$

$Re_{75}^{187} - Os_{76}^{187}$

Since these nuclides occur naturally, if unstable, they must have half-lives to the order of, or greater than, the age of the earth ($2\text{-}5 \times 10^9$ years). Such slow decay rates will be consistent with current β-decay theory only if the disintegration energy is very small or if the transition is highly forbidden or if both conditions apply. The relevence of the information which can be derived from an investigation of these nuclides, may be more clearly indicated with reference to a description of comparative half-lives and degrees of forbiddenness in β-decay theory.

1.4 Forbidden Transitions(6).

Forbidden transitions arise from the omission of two small magnitude effects in the formulation of β-decay theory. There are 1) the "source velocity effects" which are the relativistic correction terms to the vector, tensor and pseudo-vector forms of the Hamiltonian (although it is now thought that only vector and axial-vector interactions occur in β-decay the discussion is unaffected) and 2) the retardation effects which are caused by the variation of the lepton waves across the nucleus.

In allowed β-decay the rate is expressed as a function of the matrix elements M_F and M_{GT} in which the second and higher order terms were assumed negligible. When ΔJ, the change in nuclear spin from the initial to the final state, is zero and $\Delta\pi$, the change in parity is -1, the first order terms of the matrix element vanish and the velocity interactions may contribute and produce forbidden transitions. Similarly the largest retardation term in the expansion of the wave equation vanishes unless $\Delta J=0$, 1 and $\Delta\pi = -1$ and also if a transition from an initial to final nuclear state $J=0\rightarrow J=0$ occurs. Transitions governed in this manner are called first-forbidden transitions.

Applying the retardation effects to the velocity interactions in turn, yields still more highly forbidden transitions. The largest retardation effect, n=1 on the velocity term produces twice forbidden transitions and each successive retardation term raises the "degree of forbiddenness" by unity. The parity charge alternates with successive degrees of forbiddenness.

Consequently forbidden decays are more sensitive to the nature of the theory than are allowed decays.

1.5 Selection Rules[7].

Table II.

Degree of Forbiddenness	Parity Change	Spin Change	log ft
Allowed	No	$0, \pm 1$	
1st	Yes	$0, \pm 1, \pm 2$	7±1
2nd	No	$\pm 2, \pm 3$	~13
3rd	Yes	$\pm 3, \pm 4$	~18
4th	No	$\pm 4, \pm 5$	~23

1.6 The Comparative Half-Life[6,8].

λ, the probability per unit time that a nucleus will undergo β-decay is given by the formula $\lambda = \int_1^W N(W)\, dW$, where N(W) is the number of β-rays observed in the energy interval dW. This can be expressed in terms of the Fermi (M_F) and Gamow-Teller(M_{GT}) matrix elements as

$$\lambda = \frac{g^2}{2\pi^3}\left(C_F^2 / M_F^2 + C_{GT}^2 / M_{GT}^2\right) f(Z,W_o) \qquad (1)$$

where C_F and C_{GT} are the Fermi and Gamow-Teller coupling constants and

$$f(Z,W_o) = \int_1^{W_o} pW(W_o-W)^2 F(Z,W)\, dW.$$

$pW(W_o-W)^2$ is a statistical weighting factor and F(Z,W) is the coulmb correction to the spectrum shape.

For positron decay the formula in identical but a negative value of Z is used. (Normally the half-life is shorter since K-capture will complete). No naturally occurring positron emitter

/emitter has been observed yet. In electron capture, however, the coulomb factor is different since the decay rate depends on the properties of the electron cloud in the neighbourhood of the nucleus. Values for $f(Z,W_o)$ are therefore different for and electron-capture. As this does not affect the qualitative discussion being presented, no further reference will be made to this difference in the theory.

The half-life '$\underline{t}$' of a transition is given by $t = \frac{\log 2}{\lambda}$. Substituting the value for λ, from equation (1) it is apparent that '$\underline{t}$' is a widely varying function dependent on the value of the end-point energy ($\sim W_o^{-5}$). It is not, therefore, a suitable parameter for classifying the type of transition e.g. allowed, 1st forbidden, etc and can range in value from a few seconds to many years for an allowed decay. But, since the matrix elements are of the same order of magnitude for each transition, the product '$ft' = \frac{B}{(1-x)/M_F^2 + x/M_{GT}^2}$ is approximately a constant. B is a constant derived from equation (1) and '$x' = C_{GT}^2 = (1-C_F^2)$ is the fractional admixture of the respective interactions.

The product 'ft' is called the comparative half-life. Since it is almost a constant for a particular degree of forbiddenness it provides a possible method of classifying the type of transition. It can vary only because of any changes in the matrix elements.

In practice it is found that the ft values may vary by as much as a factor of 500 where as a difference of $10^3 - 10^4$ is expected between each degree of forbiddenness. Ambiguity is

/is therefore possible and hence the 'ft' value above is not sufficient to allocate the transition type. However when combined with the parity, the classification can be made uniquely since this differentiates between two degrees of forbiddenness and a factor of $\sim 10^7$ exists. Information on the spectrum shape would also help since this is very different for allowed and say, second forbidden. Parity is accurately predicted by the shell model for odd A nuclei and reasonably well for even A.

Normally the 'ft' value is expressed as a logarithm. The various classifications are listed in Table II.

A knowledge of the nuclear spin and parity change specifies the degree of forbiddenness uniquely which in turn indicates the log ft value. Experimental results generally support the classification of the ft groups although there is only one measurement of a fourth forbidden transition i.e. In-115. Two other possible fourth forbidden decays are V-50 and Cd-113. Alternatively if the spin of either the initial or final states of the nuclei is deduced or known then a measure of the ft value enables deduction of the spin of the unknown state.

The log ft value is calculated from measurement of the decay energy and the half-life but in some cases it is difficult to estimate the total disintegration energy e.g. in electron-capture transitions or in β-decay with very low specific activity Under these circumstances if the degree of forbiddenness can be deduced, assumption of the log ft permits calculation of the/

/the available energy provided the half-life is measurable. A study of these highly forbidden decays, therefore, enables the deduction of mass differences between nuclei in regions where actual masses have not yet been measured. This can provide useful information near the closed proton shell Z = 50 (In-Sn-115, Sb-Te-123) and the closed neutron shell at N = 82 (La-138).

Finally, since in all the cases listed in Table I the half-lives are known to be greater than, or the order of, the age of the earth some of the decay characteristics will be applicable to methods of radioactive dating.[9-13] e.g. K-40 is known to decay to A-40 with a half-life of 1.3×10^9 years and measurement of the ratio $^{K-40}/_{A-40}$ by atoms provides a method of calculating the age of potassium bearing rocks. For such calculations an accurate knowledge of the decay scheme is essential. Rb-Sr-87 and more recently, Re-Os-187 have also been used for this purpose.

1.7 Review of the Present Position.

(A) Triple Isobars.

1. $A_{18}^{40} - K_{18}^{40} - Ca_{18}^{40}$

K-40 decays to the first excited state (2^+) of A-40 by electron capture and to the ground state of Ca-40 by β-emission.

Two completely independent methods have been used to measure this branching ratio. The first involves assaying the amount of Argon and Calcium produced by the decay in potassium bearing minerals.[9,14,15] Such rocks must be very carefully selected as the slightest porosity may permit a leakage of argon gas which will imply a low E.C./β^- ratio. Other factors which can lead to/

/to a low result are caused by incomplete gas extraction from the rocks and losses by gas adsorption during the physical handling of the minute amount of argon produced. The results for the branching ratio obtained by this method range from 0.05 to 0.12 with an average value about 0.09. Since any errors are expected to produce a low result, the 0.12 value is more likely to be correct.

Proportional and scintillation counting techniques were applied in the second method.[16,17] The characteristic X-rays and auger electrons produced in the transition have not yet been observed as they only have a few electron volts of energy and are superimposed on the β-ray continuum. Consequently the main effort has been concentrated on observations of the γ-ray arising from the de-excitation of the A-40 first excited state. NaI(Tℓ) crystals, either directly calibrated by calculation or interpolation from isotopes of known decay schemes and efficiencies or by direct comparison with a γ-ray emitter with a similar energy and known disintegration rate. Measurement of the β-activity in geiger or proportional counters presents few difficulties. Backscattering and source absorption are the main uncertainties although the comparison method with Na-24 used by McNair et al[16] to measure the γ/β ratio surmounts these.

The counting methods are now in reasonable agreement that the branching ratio is 0.12 ± 0.01. This result agrees with the largest value obtained by the geological methods.

As this nuclide was the subject of investigation elsewhere in this laboratory[16] no experiments were attempted by the author.

2. $Ti_{22}^{50} - V_{23}^{50} - Cr_{24}^{50}$

That the odd-odd nuclide V-50 is radioactive is indicated by the mass measurements of Johnson[18] which indicate an available energy of 2.39 ± 0.12 Mev for decay to Ti-50 and 1.19 ± 0.12 Mev for decay to Cr-50.

The energy of the first excited state (2+) of Ti-50 is well established. Sinclair[19] measured this level to be at 1.595 ± 0.14 Mev by neutron excitation. This is in good agreement with the level at 1.58 ± 0.06 Mev obtained by Pieper[20] in 1952 and the observation of a 1.59 Mev state in the decay of Sc-50 by Marinaga[21].

The first excited state (2+) of Cr-50 has also been measured by Sinclair to be at 787 ± 10 Kev. Van Patter[22] has verified the existence of this level and quotes a value of 780 ± 3 Kev.

Since the ground state spin of V-50 is (6+)[23,24] it can be concluded that V-50 should decay by electron capture transitu with $\Delta I=4$, no, to the 1.59 Mev level in Ti-50 and by beta emission with $\Delta I=4$, no, to the 780 Kev level of Cr-50. Assuming each of these transitions to be fourth forbidden with lo log ft ~ 23 then half-lives of the order of 10^{16} years can be expected.

As the level in Cr-50 was not discovered until 1957, the/

/the earlier experiments were designed to observe the electron-capture branch to Ti-50 and its associated 1.59 Mev gamma-ray. Heintze[25] searched for the 4.5 Kev x-rays from a source of ferrovanadium surrounding a geiger counter and quotes a minimum half-life of 3×10^{14} years. However the source used was found to contain some contamination and also no corrections appear to have been made for the facts that only a small fraction of the x-rays could escape from a thick source (27% from a 10 mgm/cm^2 source) and the fluorescent yield is 25%. Hence the lower limit deduced from the experiment is considerably shorter than that claimed by Heintze.

Baumginger and Cohen[26] using a NaI(Tl) gamma-ray spectrometer and 500 grams of V_2O_5 obtained an excess of counts in the 1.59 Mev region which indicated a half-life of $(4.8 \pm 1.2) \times 10^{14}$ years.

The first paper on V-50 (Chapter 3) describes an experiment designed to detect the 1.59 Mev γ-rays and so confirm, or otherwise, Baumginger's results. A search was also undertaken in this work for the β-rays which were expected to involve a $\Delta I=6$ transition to the ground state of Cr-50 since the 1st excited state at 780 Kev was not known at that time. No previous attempts had been made to observe the β-rays.

The identification of the first excited state of Cr-50 at 787 Kev provided another possible method of detecting the β branch via this γ-ray. Consequently McNair,[27] using a large $4\frac{1}{2}$" x $4\frac{1}{2}$" NaI(Tl) operated with a ring of geiger counters in/

/in anti-coincidence to reduce the background, recorded the γ-spectrum from a 500 gm source of V_2O_5. The high detection efficiency of this large crystal combined with the low background makes this a very sensitive arrangement. No activity was detected and lower-limits of 8×10^{15} years and 1.2×10^{16} years were found for the half-lives of the 1.59 Mev and 0.78 Mev γ-rays respectively. As no corrections were made for the shielding effects of the source in McNair's experiment it was decided to make a new examination using a still more sensitive technique which had become available. In this method five kilograms of spectrographically pure vanaduim metal were placed round a very large sodium iodide crystal. It is estimated that a factor of twenty increased in sensitivity over previous experiments is attained with this arrangement and that any gamma activity from V-50 with a half-live $< 2 \times 10^{17}$ years should be detectable. Particular attention was paid to the accurate measurement of the background spectrum which, as will be shown, can give rise to ambiguous effects.

3. $Ba^{138}_{56} - La^{138}_{57} - Ce^{138}_{58}$

Gamma activity, attributed to be associated with the radioactivity of La-138, was first ovserved by Pringle et al[28] and subsequently three γ-rays of energies 0.535, 0.807 and 1.39 Mev were reported, the latter being considered to be a cross-over transition. It was also proposed that the first and second/

/second excited states of Ba-138 were 0.807 and 1.39 Mev respectively above the ground state. However, the existence of a level at 0.807 Mev is not supported by subsequent investigation of the β-decay of Cs-138.[30] The possibility of electron capture to both these excited states was indicated and β^+ decay to either of the excited states or to the ground state was not excluded. Neither γ-γ or K-γ coincidence experiments were attempted. A half-life of 2.0×10^{11} years was suggested.

In a rather insensitive experiment, Pringle et al[29] assigned an upper limit of 12 counts/min/gram of La_2O_3 to the number of electrons or positrons emitted with energy > 100 Kev. With the aid of a large proportional counter, Mulholland and Kohman[31] reported β-emission of maximum energy (1.0±0.2) Mev, measured by aluminium absorption methods, and a partial half-life of 1.2×10^{12} years. In view of the difficulties caused by differing contamination of their absorbers, this result seemed somewhat uncertain.

A complete investigation was undertaken in the present work in an attempt to establish a decay scheme by detection of the γ and β radiation and by coincidence experiments. In parallel with this study, Turchinetz and Pringle (August 1956, Phys. Rev. 103, 1000) re-examined the γ-spectrum of La-138 and made some coincidence measurements. Their results and those describes in Chapter 3 are in good agreement.

4. $Yb_{70}^{176} - Lu_{71}^{176} - Hb_{72}^{176}$

Recent investigations[32,33,34] have conslusively confirmed the β-decay scheme of naturally radioactive Lu-176 first suggested by Gold haber and Hill[35] on the basis of unpublished work by Scharff-Goldhaber.[36] Since Lu-176 is an odd-odd nuclide, electron capture decay may also be expected but it was not observed by Scharff-Goldhaber or by Arnold[32] who concluded that the intensity is <10% of the β-rays. From an examination of the L x-ray peak, Dixon et al[34] tentatively proposed that 3±1% of the disintegrations took place by electron-capture. Since the ground state spin of Lu-176 is 10, it is unlikely that electron-capture can occur to the ground state of Yb-176 which has zero spin. Hence a search was undertaken for γ-rays or conversion electrons which could be attributed to an electron-capture branch.

$Hf_{72}^{180} - Ta_{73}^{180} - W_{74}^{180}$

The existence of Ta-180, with very low abundance (0.0123%) in natural tantalum was only recently discovered (1955) by White et al[37,38] using a two stage mass spectrometer. Calculations from the semi-empirical mass equation indicate that Ta-180 has 1.7 Mev availabe energy for electron capture decay to Hf-180 and 0.2 Mev for β decay to W-180. Eberhardt et al[39] conclude that the lower-limit half-life is 10^{13} years in the only search for activity made so far.

It is intended to examine the γ-spectrum from this isotope in the near future.

(B) Double Isobars.

1. Rb-Sr-87.

Rb-87 decays by β -emission to Sr-87. The transition is 3rd forbidden non-unique with $\Delta I=3$, yes.

Libby and Lee[40] showed Rb-87 to be the active isotope in 1939. Eklund[41] using an absorption technique to measure the β-activity concluded the spectrum was pure and there were no associated γ-rays. This was later verified by McGregor and Wiedenbeck[42] who proved by coincidence methods that the transition is to the ground state of Sr-87. There was considerable doubt, however, about the value of the β-ray maximum energy which was allocated values varying between 130-560 Kev. Later work by Curran[43] using a large proportional counter, Lewis[44] with a rubidium iodide crystal, and McGregor[45] by β -ray spectrometry, agreed that E_β = 275 Kev. This has since been confirmed by Flynn and Glendenin.[46]

Particular interest is concentrated on the use of Rb-87 as a method of dating ancient minerals and rocks. To do this an accurate knowledge of the half-life is necessary and present work has been guided towards this end. Results for the half-life obtained by applying chemical and mass spectrographic methods to determine the $^{Rb}/_{Sr}$ ratio indicate a consistent value of $\sim 5 \times 10^{10}$ years.[47] When this value is used in calculating the ages of rocks good agreement with the results given by the Uranium-Lead and Potassium-Argon dating is attained.

When counting methods are used the half-life is found to vary between 4.3 and 6.4 x 10^{10} years. Curran[43] has found that the beta spectrum measured in a proportional counter with a solid source has no maximum, there being a very large number of low energy electrons whilst McGregor and Wiedenbeck,[45] in a thin lens β-ray spectrometer, find that this spectrum produces a linear Kurie plot when the third forbidden Vector interaction correction factor is applied. It is a consequence of the soft electron spectrum that the normal corrections to the counting techniques e.g. solid angle, source self-absorption, back-scattering and bias level, become difficult to assess accurately.

A very recent measurement by McNair,[48] using a 4π counter with an enriched rubidium source aims at minimimising these uncertainties. He finds the half-life to be 5.25 x 10^{10} years which is in fair agreement with the value accepted by the geologists.

No further work was attempted on this isotope as the half-life and maximum β energy seem to be well-established now.

2. Cd-In-113.

The decay systematics of Wapstra[49] predict that Cd-113 should be the active isotope with an available energy of 150 Kev for β decay to In-115. Calculations[50] from the table of empirical atomic masses[5] also indicate that Cd-113 is β labile but with an available energy of 430 Kev.

No attempts have been made to observe the β activity/

/ activity although Cohen[51] using a krypton filled proportional counter, searched for the Cd K x-rays which would arise in the event of K electron capture in In-113. No x-rays were observed and he concluded that the half-life of In-113 for K capture is $>10^{14}$ years. Some L x-rays were detected indicating a half-life of the order of 10^{12} years for L capture. However it is likely that the greater part of this radiation is due to the interaction of the cosmic ray background with the source as first observed by Dixon and McNair.[52] Heintze,[25] using a large proportional counter was also unsuccessful in an attempt to detect K x-radiation from In-113 and concluded that the half-life $>10^{14}$ years. The failure to detect K x-radiation prompted the present author to examine Cd-113 for β activity.

3. In-Sn-115.

Riddell's mass differences[50] predict that In-115 will decay to Sn-115 with a maximum β-energy of 270 Kev. Martell and Libby,[53] using a screen-walled geiger, detected β radiation from In-115. They estimated from absorption measurements that the end-point energy was 630 Kev and the half-life was 6.0×10^{14} years. The use of aluminium absorption foils in very low specific activity measurements is always doubtful because of possibly varying radioactive impurity levels. Cohen[51] later confirmed that In-115 was radioactive and that the beta radiation was 'a few hundred Kev'. The work of Varma and Mandeville[54] and Gorodetzky et al[5] on the metastable states/

/states of In-115 infers that the beta energy of the In-115 ground state should be nearer 500 Kev. As there is some doubt about the value of the end-point energy a new measurement was undertaken.

4. Sb-Te-123.

It cannot be deduced from the energy systematics which is the unstable isotope. The empirical mass difference tables[50] predict that Sb-123 has 84 Kev available energy for beta decay to Te-123 where as Wapstra[49] predicts that Te-123 should be the active isotope, with a Q value of 186 Kev.

Meintze[25] examined natural tellurium for possible x-rays in a proportional counter and concluded that the half-life of Te-123 for K capture is $\geq 10^{13}$ years. As no activity has been observed in either isotope it was decided to search for possible beta, K-capture and gamma activity in a more sensitive experimental arrangement.

5. Re-Os-187.

In 1948 Naldrett and Libby[56] claimed to have detected the radioactivity of rhenium in a screen-walled geiger counter. They concluded from absorption measurements that the β-radiation had a maximum energy of 43 Kev with a half-life of $\sim 4 \times 10^{12}$ years. Gauthe and Blum[57] using nuclear emulsion techniques also concluded that rhenium was beta-active but that the energy was only 11 Kev. This result prompted a re-investigation of the rhenium beta end-point energy by Suttle and Libby[58] who found that the energy was < 8 Kev and the half-life $\leq 10^{11}$ years.

They were unable to obtain an accurate half-life determination because of the serious absorption of the very soft beta-radiation in the solid source. It appears that Libby's earlier result was caused by contamination and the discrepancy between the photographic and counting technique is due to the difficulty in accurately calibrating the emulsion. In an attempt to resolve the uncertainty about the rhenium decay energy, Dixon and McNair[52] examined a rhenium source in a large proportional counter and concluded that there were no beta-particles with energy > 1 Kev to put a lower limit of 10^{16} years on the half-life. A search for possible K and L-capture in Os-187 was also unsuccessful although some K and L x-rays observed were proved to originate from the actions of the cosmic radiation on the source. They concluded that Os-187 was stable with a half-life > 1.4×10^{16} years. That Re-187 is the active member of the isobaric doublet seems to have been established by the strong evidence of Hintenberger et al[59] and Herr et al[60] who have observed abnormal abundances of Os-187 in geologically old, rhenium containing molybdenites and have recently applied the method to dating this type of rock. A half-life of 6.2×10^{10} years is deduced from this work.

Owing to the apparently short half-life of Re-187, if the beta-radiation is 1 Kev it should have been easily detectable with existing counting techniques although it is possible that most of the soft electrons are being lost either byattachment to small amounts of vapour arising from the adhesive material/

/material used to mount the source or more likely, due to electrostatic charging of the rhenium oxide used in the counting experiments. A loss of counts due to improperly conducting sources has frequently been experienced by workers in the 4 counting field. The effect becomes progressively serious with softer beta-emitters. It was thought that these difficulties could be avoided by searching for the beta radiation with an evaporated metallic rhenium source in the counter and examining the spectrum to very low energies (100 ev).

Chapter 1.

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Chapter 2.

The Attainment of Low Backgrounds.

It is implicit in the previous discussion that, if radioactive, the nuclides scheduled for investigation must have very low specific activities. Hence, to achieve the maximum sensitivity of the counting equipment, the lowest possible backgrounds combined with the largest practicable source quantities are essential. To reduce the background consider the following points.

2.1 Sources of Background.

Ionising radiation, produced directly or indirectly by the interaction of the cosmic radiation with the detector is the fundamental source of background in counters. Other effects contributing to the background, e.g. local radioactivity in the surroundings or construction materials, and spurious pulses in the electronic system, are of a secondary nature and can be avoided by careful construction of the counting equipment and selection of pure materials. Hence a low background system designed such that cosmic radiation will have a minimum effect will be the ultimate - assuming the secondary effects are, or can be, removed. Consideration of results of experimental work on the nature of the cosmic radiation indicate the most effective methods of reducing this component of counter background, which at sea level is mainly produced by the extensive air shower. The mechanism by which this is created is fully described by Galbraith(1).

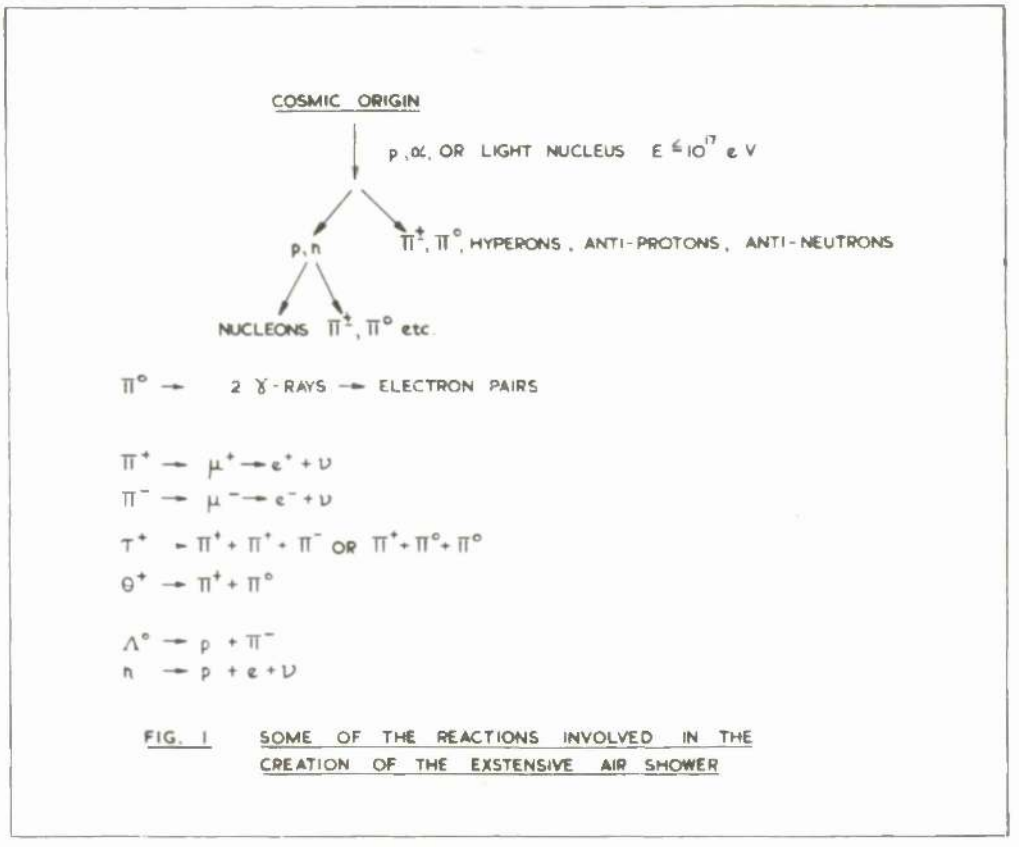

FIG. 1 SOME OF THE REACTIONS INVOLVED IN THE CREATION OF THE EXSTENSIVE AIR SHOWER

2.2 The Extensive Air Shower.

The primary radiation, mainly of very high energy protons ($\leq 10^{17}$ eV) and alpha particles with a small amount of heavier nuclei, interacts with the oxygen and nitrogen at a great height in the atmosphere to produce a shower of nucleons, charged and uncharged π mesons, and probably hyperons and anti-particles. This forms the 'core' of the shower which travels along the direction of the original particle and spreads over about one square metre area at sea level. The charged mesons may undergo spontaneous decay into a μ-meson, which forms a penetrating non-interacting component, or may interact in a similar manner to the fast protons - the mode of behaviour depending on the energy of the π meson. The π° mesons decay almost instantaneously into two gamma-quanta, which in turn produce the photon-electron cascade. This can extend over 500 square metres at sea level.

Some of the core particles, produce further nuclear disintegrations yielding more mesons and nucleons to form the nucleon cascade.

To summarise, the extensive air shower (Fig. 1) is a core of high energy particles some of which are nuclear interacting (N-component) around which are distributed the photon-electron cascade including μ-mesons. The N-component (neutrons, protons and π mesons), and the μ-mesons are highly penetrating. At sea level the μ-mesons constitute about 10% of all shower particles. The ratio of nucleons with π and μ-mesons, to all particles is/

/is about 0.3 and the number of neutrons present is 1-2% of the electron density, i.e. of the order of 10^4 neutrons at sea level/ 10^{16} eV primary.

Coulomb scattering spreads the secondary component of the shower over a large area, the density of particles falling off with the distance from the core. With a shower produced by a lower energy primary, most of the components may be absorbed in the atmosphere leaving only the μ-meson component.

Experiments on delayed particles caused by the different speeds of electrons, μ-mesons, the N-component and different path lengths due to scattering and inclination to the shower axis, show that more than half the particles arrive within 10^{-7} secs. The estimated fraction of delayed counts is 0.85% of the shower particles and these have delays $< 10^{-6}$ secs. Since all the components are time coincident within 1 μ sec. it follows that if any one of the particles or photons associated with a shower are detected then the contribution, of any part of this shower, to the background of a counter can be removed by anti-coincidence techniques. However, since the electron-photon cascade is much more extansive than the core of the shower, the relatively soft electrons can be absorbed in the atmosphere leaving only the pnotons, which may interact in the counter without recording in the anti-coincidence assembly. Background from this cause can be reduced by using anti-coincidence counters with a high gamma-efficiency, e.g. scintillation counters or alternatively by providing suitable shielding to absorb the photons. Greisen(2)/

/Greisen (2) has calculated the probability of a cascade particle being detected under large thicknesses of lead. Experimental verification of his results show that 8" of lead effectively absorbs the photon-electron cascade in shower phenomenon. Cocconi (3) has investigated the mean free path of the primary component in materials of different atomic numbers and finds that it is higher in lead than in carbon. This is because the primaries interact with a cross-section close to the geometrical and since in lead the number of nucleons screened by outer nucleons is greater than for light nuclei, the heavy nuclei have thus a smaller stopping power/nucleon.

2.3 Neutron Production.

Neutron production caused by the introduction of massive shielding in the path of the cosmic radiation has been investigated by Tongiorgi (4-6) who concludes that the interaction of the N-component in lead absorbers produces a large number of 10 - 20 MeV neutrons in penetrating showers. The energies of these ne neutrons are characteristic of evaporation neutrons and since they are produced in the lower 5 cm. of lead absorbers, cannot be from (gamma,n) reactions but are due to the interactions of highly penetrating neutron, protons, or π and μ mesons. About 60 neutrons can be produced by a single high energy nucleon interacting in the lead to produce a penetrating shower of mesons and nucleons which give rise to further neutrons in the lead absorber. Cocconi (7) finds the average neutron multiplicity to be ten times. These are distributed isotropically. 40 cm. thickness/

/thickness of paraffin wax is sufficient to thermalise these neutrons which take about 200 μ sec. to slow down. This time delay must be allowed for in any anti-coincidence systems designed to remove the effect of the neutron component in showers produced in the shielding.

Significant results on neutron shielding were obtained from Tongiorgi's experiments. The neutron count rate under 5 cm. of lead shield is only slightly reduced by the addition of 25 cm. thick paraffin wax outside the lead. Removal of the lead greatly reduces the neutron production whilst addition of lead outside the paraffin wax increased the rate slightly. Insertion of a further lead layer between the counter and the paraffin wax greatly increases the neutron production rate again. In addition the neutron yield is found to increase with the atomic number of the absorber. It follows that the most effective neutron shield should be made from material of low atomic number e.g. paraffin wax and there must be no high Z material between the sensitive volume of the counter and the neutron shield (8).

Examination of cloud chamber photographs (9) show that cascades can be initiated in absorbers penetrated by photons, neutrons and π° mesons without associated charged particles. If the anti-coincidence system was placed outside the shielding material it would not detect most of the cascades produced by these uncharged particles.

2.4 The Ideal Theoretical Cosmic-ray Shield for Radiation Detectors.

It is apparent that the presence of any abosrbing material in the path of a primary cosmic ray can lead to the production of a wide range of charged and uncharged particles, which, with the possible exception of the neutron component, are time coincident to within 1 μ sec. In practice it is clearly impossible to remove all absorbing material from a wide enough area surrounding the radiation detector to ensure that particles undergoing multiple coulmb scattering, produced as decay products, will not be directed into the counter and recorded. Consequently the first step in building a system with zero background for cosmic rays is to provide a system of counters completely surrounding, and operated in anti-coincidence with the detector. This will remove the largest effect of the cosmic radiation but does not allow for the possible interaction of photons from the electron-photon cascade where the electrons may be absorbed out and the remaining photons pass undetected through the anti-coincidence counters to record in the detector. This effect can be reduced by having an anti-coincidence system with a high gamma-ray efficiency, but as 100% efficiencies are unobtainable shielding is necessary to reduce the gamma-intensity. Greisen finds that 8" of lead is sufficient for this. Counts due to the production of showers in the shielding are largely removed if the anti-coincidence system is placed inside the shield since there is a very high probability that an associated /

/associated charged particle from the cascade will be detected. There is still the possibility, however, that uncharged particles scattered from the surroundings or produced in the immediate shielding will reach the detector without triggering the anti-coincidence arrangement.

Tongiorgi has proved that the best method of removing the contribution from the uncharged particles is to have about 16" thick paraffin wax under the gamma-ray shield. If this wax is impregnated with a material having a high neutron capture cross-section and which does not produce capture gamma-rays, then there should be a very low neutron transmission through the wax. Boron admirably fits this requirement as it has a high capture cross-section for thermal neutrons and the alpha particles emitted in the reaction are readily absorbed. This shield has a low atomic number and is less likely to initiate showers. The thermalising process of the neutrons takes approximately 200 μ sec. so the anti-coincidence system must be designed to remove coincidences with the occurrence of delayed events in this period.

As cascade events can also be initated by interaction of fast particles in low Z materials, although with smaller probability, the anti-coincidence system should be placed under the neutron absorber where there is a good chance they will be detected.

2.5 Experimental.

Ramsden and Watt have conducted a series of experiments designed to find the most efficient position in the shielding arrangement for the anti-coincidence counters.

A small proportional counter was surrounded by a ring of geiger counters and placed inside a 4 inch thick steel shield which, in turn, was covered by a second layer of geiger counters. The results are shown in Table I.

Table I.

Proportional counter	c/min.
1. Inside 4" Fe shield	17.3 ± 0.3
2. In shield with outer geigers operating in anti-coincidence	7.7 ± 0.26
3. In shield with inner geigers operating in anti-coincidence	6.5 ± 0.2.
4. In shield with both rings operating in anti-coincidence	6.0 ± 0.1

It is apparent that the optimum position for the anti-coincidence ring is inside the shield as predicted in the foregoing discussion. The increased efficiency when both sets of geiger counters are used is caused by the dead spaces between any two counters and also to the low gamma-ray efficiency. These conclusions have been confirmed by operating a ring of geiger counters in anti-coincidence with a wall-less counter of the Drever (10) type in which the built-in anti-coincidence system has no dead spaces.

Assuming no spurious pulses are produced in the electronic/

/electronic equipment, the residual background count rate observed in a wall-less proportional counter is entirely due to uncharged particles and/or gamma-radiation interacting within the sensitive volume of the counter. The effect of charged particles in the cosmic radiation and radioactive impurities in the construction materials are removed very efficiently by the internal anti-coincidence arrangement. The instrument is therefore ideally suited for resolving the components of background due to gamma-radiation and to neutron interaction.

The large 24 litre (12 litre sensitive volume) wall-less counter normally used for radio-carbon dating (11) was placed inside the water-shielded cell (12) at Aldermaston and the anti-coincidence count rate recorded above 500 eV energy. Bricks made from 55.2% paraffin, 36.8% polythene and 8.0% boric oxide each 20 cm. thick were placed around the counter to ensure thermalisation and capture of any neutrons in the background.

Table II shows that a factor of two reduction in background attributed mainly to removal of neutron component and partly to γ-ray shielding, was obtained leaving a residual count rate of 4.1 ± 0.08 c/min. Increasing the pressure of gas in the counter inside the wax shield resulted in a rise in count rate which could be caused by

(1) inefficient neutron shielding by the wax

(2) increased γ-ray detection

(3), radioactive impurities in the counting gas.

To resolve the contributions, a 0.020" thick layer of cadmium/

/cadmium metal was then placed between the counter and the wax bricks. Since cadmium has a very high thermal neutron capture cross-section and produces several γ-rays in the reactions, if there were any neutrons leaking through the shield the count rate would increase, for calculation shows the γ-ray efficiency of the counter to be ten times the neutron efficiency. The 12% reduction in count rate using cadmium inside the boron-loaded wax shield is consistent with the attenuation expected for 510 KeV γ-rays. It is concluded therefore that the neutron leakage must be very small. Similarly any activity from impurities inside the counter or spurious pulses arising in the electronics must be negligible and the residual background with cadmium and polythene shielding is almost entirely caused by external γ-rays.

Table II.
Wall-less Counter in Low Background Cell.

Additional Shielding	c/min.
(Gas Pressure 59 cm Argon + 10 cm CH_4)	
None	8.1 ± 0.16
12 inch thick boron-loaded wax	4.1 ± 0.08
(Gas Pressure 105 cm Argon + 10 cm CH_4)	
12 inch thick boron-loaded wax	8.8 ± 0.18
0.020 inch cadmium foil + wax	7.8 ± 0.16

De Vries (13,14,15) has made a careful investigation of the contribution from neutrons to the background of counters and concluded that neutrons are produced in the top layer of the steel shield. Addition of a 15 cm layer of boron-loaded wax/

/wax placed in the steel shield reduced the neutron component by a factor of 7. He obtained the lowest background when a polythene sleeve surrounding the counter was removed. From the results of the present work it is concluded that the polythene used in De Vries experiment was thermalising the neutrons produced in the layer of steel between the boron-loaded wax and the counter thus incressing the probability of detection or, alternatively, the polythene sleeve was contaminated. A 2% fluctuation in the meson intensity per cm. Hg. change in barometric pressure was observed by De.Vries. For a completely efficient cosmic ray shield however, this variation should not matter.

2.6 Conclusions.

Results from cosmic ray investigations and counter background experiments lead to the following conclusions on shielding.

6.1 The detector should be completely surrounded by a system of counters which has 100% efficiency for the detection of particles and as high a gamma-ray efficiency as possible. Geiger counters and proportional counters are most frequently used but plastic phosphor counters or an anulus of sodium iodide crystal would be better because of their higher gamma-ray efficiency. It is best that the detector pulse be delayed by at least 200 μ sec. and the anti-coincidence pulses made sufficiently long to prevent recording possible events caused by the delayed neutrons.

6.2 About 16" thick boron-loaded polythene or paraffin wax placed/

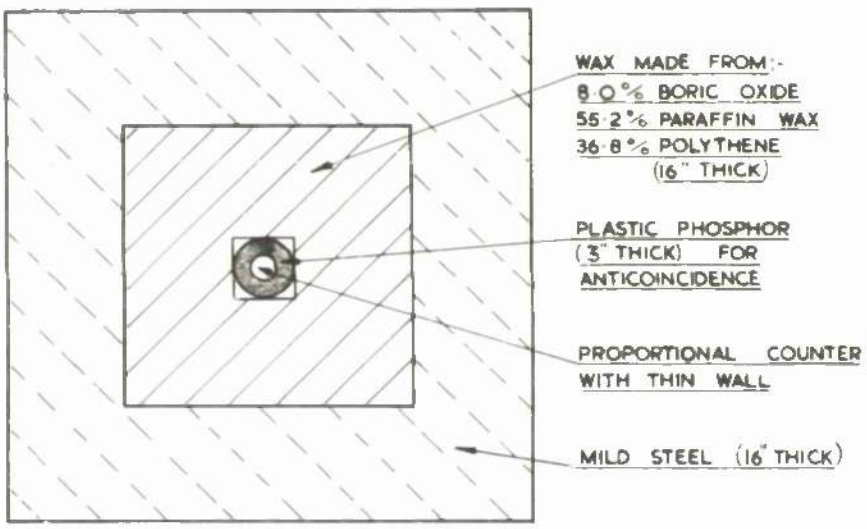

FIG. 2 OPTIMUM SHIELDING ARRANGEMENT

/round the anti-coincidence counters will effectively thermalise and remove the neutron component. The presence of the neutron capture material reduces the necessity for the 200 μs delay on the gate. There must be no appreciable thickness of high atomic number material between the detector and the wax otherwise cascades will be initiated and more neutrons produced as observed by Tongiorgi.

6.3 Finally, a layer of lead about 8" thick, or material of equivalent stopping power for gamma-rays, is required outside the neutron shield to remove the photons (510 KeV) associated with the electron-photon cascade. A low Z material may be a slightly better shield because the range of the penetrating particles is smaller and fewer neutrons are likely to be produced.

Figure (2) is a diagrammatical expression of the arrangement for removal of the background contribution from cosmic rays.

Having decided upon the most efficient method of reducing the background contribution from the cosmic rays one has potentially the lowest background attainable - the limit being determined by the degree to which the remaining factors listed in Section 2.1 occur. These factors, together with suggestions on how they can be reduced, are considered below.

2.7 Local Radioactivity.

Radiation from the surroundings or construction materials/

/materials of a low background counting assembly may arise in several ways.

1. Natural Radioactivity.

Elements with naturally radioactive isotopes occur in nature. By definition these have very long half-lives, approximately the age of the earth, but short-lived isotopes can occur in the radioactive decay chain in equilibrium with the long-lived parent. The natural radioactive chains of Uranium, thorium and actinium are typical of this class and are the most commonly experienced, being found in small but measurable quantities in nearly all materials. K-40, the radioactive isotope of potassium is abundant in nature and is nearly always found in brickwork, cement and glass and all common building materials. C-14 is constantly produced by the action of the cosmic rays in the atmosphere. Although it emits soft beta-particles which are easily absorbed, the resulting bremsstrahlung may record in the counting system. Less important natural activities are lanthanum (La-138), lutecium (Lu-176), indium (In-115) and rubidium (Rb-87). The remaining naturally radioactive isotopes are unlikely to be troublesome.

Careful selection and testing of materials used for the counter construction and shielding is the only method of avoiding this source of background short of actual chemical separation of the active material. The results of tests for activity on specific materials have been published by several workers but these vary a great deal according to the source of supply. It/

/It is generally agreed that electrolytic copper is usually very pure. Lead free brass is also a suitable construction material.

The author finds that when both brass and copper are used for proportional counters adsorbed layers of radon or thorn on the surface increase the background. Activity from this effect can be reduced if the counters are treated by the method described in Section 3. In scintillation counting, potassium present in the glass of the photomultipliers and sodium iodide crystals was at one time the main source of activity and easily identified by the characteristic gamma-peak at 1.46 MeV. Manufacturers have been very successful in reducing the presence of this material to negligible proportions. It is our experience that gamma-radiation observed in a scintillation counter spectrum associated with K-40 arises from the surrounding building materials and can be reduced to negligible proportions with adequate shielding. Mild steel is a suitable shielding material as it has a very low potassium and uranium-thorium content.

A sensitive method of determining whether activity is arising in the shielding and materials outside the actual counter is to place a gamma-ray detector inside the shield and record the spectrum. Then place a known layer of pure material, such as electrolytic copper, round the detector and record the spectrum again. Any peaks appearing in the difference between the air and the copper spectrum must be caused by external radiation. The method depends on the fact that gamma-radiation

undergoes/

/undergoes the normal exponential absorption whereas the cosmic ray background is relatively unaffected. (See 'The Half-Life of V-50 in Chapter 3).

A frequently employed method of reducing the effect of gamma rays arising from external radioactive impurities is to place a layer of triply distilled mercury between the detector and the anti-coincidence ring. Whilst an improvement in count rate has been reported that method destroys the efficiency of the cosmic ray shield and prevents attainment of the ultimate in background. The improved count rate simply means there is a relatively high activity in the shield or surroundings. Glass geiger counters, commonly employed as the anti-coincidence detectors, should be avoided as they usually contain K-40. Metal counters are not so prone to this defect and are better but the best system is to have the anti-coincidence system built-in, e.g. Drever et al. Ramsden.

Bremsstrahlung arising from C-14 activity is easily absorbed out in the shielding but care must be taken to avoid having materials made from modern carbon inside the shield.

2. Induced Radioactivity.

Other possible factors affecting the radioactive level in the surroundings of the counter system are caused by fission products in the radioactive fall-out from nuclear weapons tests, A-41 and neutrons from reactors, large accelerating machines in the vicinity, also airborne activity such as radium, thorium and tritium.

Careful siting of the counting laboratories in a position remote from reactors and machines avoids this source of radiation. Testing and selection of construction materials is necessary to ensure the absence of fission products. Ferguson flushes his equipment continuously with Nitrogen gas to remove the effects of adsorbed radioactive gases. The other alternative is to have a sealed laboratory with a closed cycle ventillation system under which the shorter-lived activities will soon decay.

3. Construction Materials.

Freedom from radioactive materials in the construction of the detector and its immediate attachments, including the anti-coincidence assembly, is of the utmost importance in the achievement of very low backgrounds. Experiments show that considerable activity can arise from K-40 and the natural U-Th series in glass geiger counters and photomultipliers, and these should be avoided where possible. Recently manufacturers have been building photomultipliers with potassium-free glass specially suited for low background applications. If geiger counters are used for anti-coincidence work they should be made from brass or copper as these are usually of high purity.

Proportional counters constructed from lead-free brass or electrolytic copper with quartz or ebonite insulators have been found to give good results although some activity has been traced to adsorbed active gases e.g. (Rn,Tn) on the walls of the counter. Purging with nitric acid and distilled water greatly reduces this activity especially if the counter is never opened directly/

/directly to the atmosphere. If the background count rate of a detector, kept out of contact with the atmosphere, falls off with time, one can assume this is caused by the decay of adsorbed activity - usually of the four day radon.

The quenching agents used im most porportional counters contain modern carbon which contributes to the count rate. It is possible to obtain such gases as methane, acetylene, CO_2, made from old carbon thus removing another source of background.

The effect of radioactive impurities in the construction material of proportional counters can be largely removed by building the wall-less type of counter. As this counter has a built-in anti-coincidence system any counts from particle activity in the walls are vetoed.

Miller (16) has analysed the spectra obtained with sodium iodide crystals recording coincidences and anti-coincidences with cosmic ray counters surrounding the crystals. He concludes that there is Ra-226 in the aluminium casing and that there is K-40 in the phototubes, crystal and canning windows. When the crystal was canned in stainless steel with a quartz window a factor of two reduction in background was obtained. The final background being 150 c/min. $>$ 90 KeV under 8" thick steel for a 4" x 1½" NaI crystal. Alpha-activity observed from the materials confirmed the presence of U-Th series as commonly experienced by the author. Aluminium has been found to be a bad material for low background work confirming Andersons (17) results. A crystal with electrolytic copper can and quartz window was found/

/found to give 18 c/min/cub. ins. compared with 24 C/min/cub. ins. for an aluminium crystal under 20" steel and with the bias level at 30 KeV.

Attempts to reduce the background in crystals by using cosmic ray counters in anti-coincidence have not been as successful as have those with proportional counters. Both geiger counter and scintillation counter anti-coincidence assemblies reduce the background of the detector by only about 30%. The neutron contribution to the count rate of a crystal can be obtained using Brook's (18) method of rise time differences in anthracene or alternatively to shield the crystal with boron-loaded polythene and operate it with an anti-coincidence anulus.

2.8 Shielded Laboratories.

The elaborate shielding arrangements outlined in section 2.6 was not available for the experiments described here. However, the general principles pertaining to low backgrounds were applied by utilising an underground site at Glasgow University and the special low background facilities at A.W.R.E. Aldermaston. In these laboratories the best counter backgrounds so far attained by other workers in a study of the double and triple isobars were surpassed.

1. The Underground Laboratory at Glasgow University.

In an attempt to reduce the cosmic ray background a laboratory was fitted out in a disused coalmine which runs under this University. The laboratory was 90 feet underground and test measurements made with a geiger counter indicated that the/

/the particle component was reduced by a factor of three on the surface intensity. However, the background for a 2" diameter NaI(Tℓ) crystal recorded inside and outside a 4" steel shield in the underground laboratory showed a factor of twenty difference. This is interpreted to be due to a γ-flux present in the surroundings of the mine. Also the background of the crystal on the surface has only a slight slope when additional layers of lead are placed over a 4" lead shield whereas in the mine the slope is much steeper. As an additional check rock samples were taken from the surroundings and count rates recorded. Table 4 shows the results which indicate that a shale bed on the roof of the laboratory is the primary cause of the activity with smaller contributions from the mineral vein in one wall and from the surrounding brickwork. Despite the disadvantage of this local contamination (later shown to be due to the U-238, U-235 and Th-232 naturally radioactive series) it was found that with the lead shielding increased to 8" thick most of the γ-radiation was removed and a very good background obtained.

TABLE 4.

Background of crystal > 63 Kev = 36 c/min.

Sample	Counts per minute per 210 gms. (to 5% statistical accuracy)
Brick	50
Cement	36
Mineral Vein	40
Shale	77
Sandstone	30

The count rate below background from the sandstone is caused by the self-shielding effect of the source.

Two methods of pulse analysis were used. In the first method pulses from the detector were amplified, displayed on an oscillograph and recorded on 35 mm film with a camera geared to use 100 feet of film in 12 hours. This procedure has the advantage in that any spurious pulses are easily recognisable and can be discarded also in the event of breakdown during a run some of the results can be salvaged. On the other hand, analysis of the results is performed manually on a projector - a very tedious time - consuming task as an average run of 12 hours requires about 48 hours analysis.

A method was developed for transmitting the pulses along 400 feet of cable, using a booster cathode follower, to a pulse height analyser situated at ground level.

2. The Water-Shielded Laboratory at A.W.R.E.

This laboratory [12] is a circular room 18 feet in diameter constructed out of 1 inch thick mild steel plate and surrounded by an outer shell which permits 6 feet of water shielding round the sides and 7 feet on top. The door is 12 inches thick steel. A principle applied to the construction was to exclude any possible path which did not pass through at least one foot of steel or its equivalent. All paints and construction materials were tested for activity with geiger and scintillation counters and only the materials with no observable activity were used.

No electronic equipment other than pre-amplifiers were permitted inside the cell, the signal and power cables being led through the tank wall to the ordinary adjacent laboratory where the necessary electronics were stored.

Results on background experiments with proportional and scintillation counters show this low background laboratory is a better shield than 12" of steel shielding in the brick laboratory.

Chapter 2.

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Chapter 3.

The Triple Isobars (La-138. V-50 and Lu-176).

3.1 The Natural Radioactivity of Lanthanum.

An investigation, using a proportional counter, into the reported β activity of La-138 and an attempt to establish the decay scheme by means of γ-ray coincidence methods, using scintillation counters, was conducted in the underground laboratory at Glasgow.

1. Experimental Arrangements.

The scintillation spectrometer consisted of a 2 inch high x 2 inch diameter NaI crystal mounted on a DuMont 6292 photomultiplier and surrounded on all sides by 8 inch lead. The amplified output pulses were fed into a cable 400 feet long and analysed at the surface by means of a 100-channel Hutchinson-Scarrott kicksorter. The total background above 70 Kev was reduced to 80 counts per minute compared with 250 c.p.m. at ground level.

Coincidence experiments were carried out under the same shielding conditions, the second detector being a NaI crystal of the same size and identically mounted.

For the detection of β-rays the system used was basically that described by Dixon and McNair [1]. The proportional tube spectrometer was surrounded by two rings of Geiger counters and shielded by 4 inch lead and 1 inch iron on all sides. The proportional counter, diameter 14.4 cm, has been modified so that

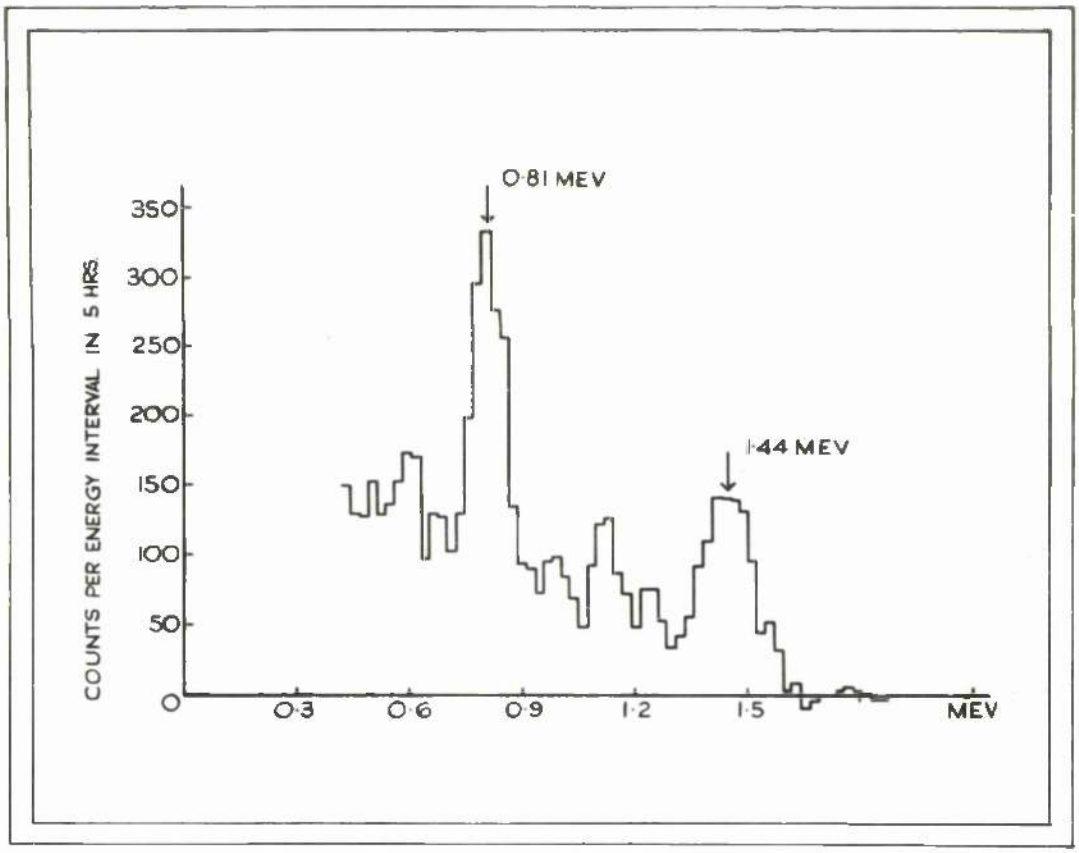

0·81 MEV
1·44 MEV
COUNTS PER ENERGY INTERVAL IN 5 HRS.
350
300
250
200
150
100
50
0
0·3
0·6
0·9
1·2
1·5
MEV

FIG 1.

/that 32.1 cm of its cathode is fully operative, giving an available internal cathode area of 1450 cm^2, an increase of roughly 30%. As previously described, the amplified pulses were displayed oscillographically and filmed. With the counter at a pressure of 4 atm. (argon-methane mixture), the background above 20 Kev was 27 c.p.m. as compared with a value of 50 c.p.m. at ground level, and 30 c.p.m. is the basement site used by Dixon and McNair (1954) some 10 feet below ground level. The reason for the rather disappointing decrease is the increased low energy γ-ray flux, mainly due to ^{40}K, which cannot be completely absorbed by the available shielding.

2. Gamma-Ray Spectrum.

The source consisted of 7 g of La_2O_3, distributed uniformly round the crystal, the average source thickness being approximately 70 mg/cm^2. The activity due to the source was 19 c.p.m. against a background of 49 c.p.m. The spectrum with background subtracted (Fig. 1) shows the presence of two γ-rays of energies (0.81 ± 0.01) and (1.44 ± 0.02) Mev. No other γ-ray was observed with energy < 2.5 Mev. In particular, closer investigation failed to reveal either annihilation radiation, which would be expected if β^+-emission occurred, or a γ-ray of energy 0.535 Mev. It seems probable that statistical fluctuations caused Pringle et al[2] to interpret the Compton edge of the 0.81 Mev -ray as evidence for the existence of a 0.535 Mev γ-ray.

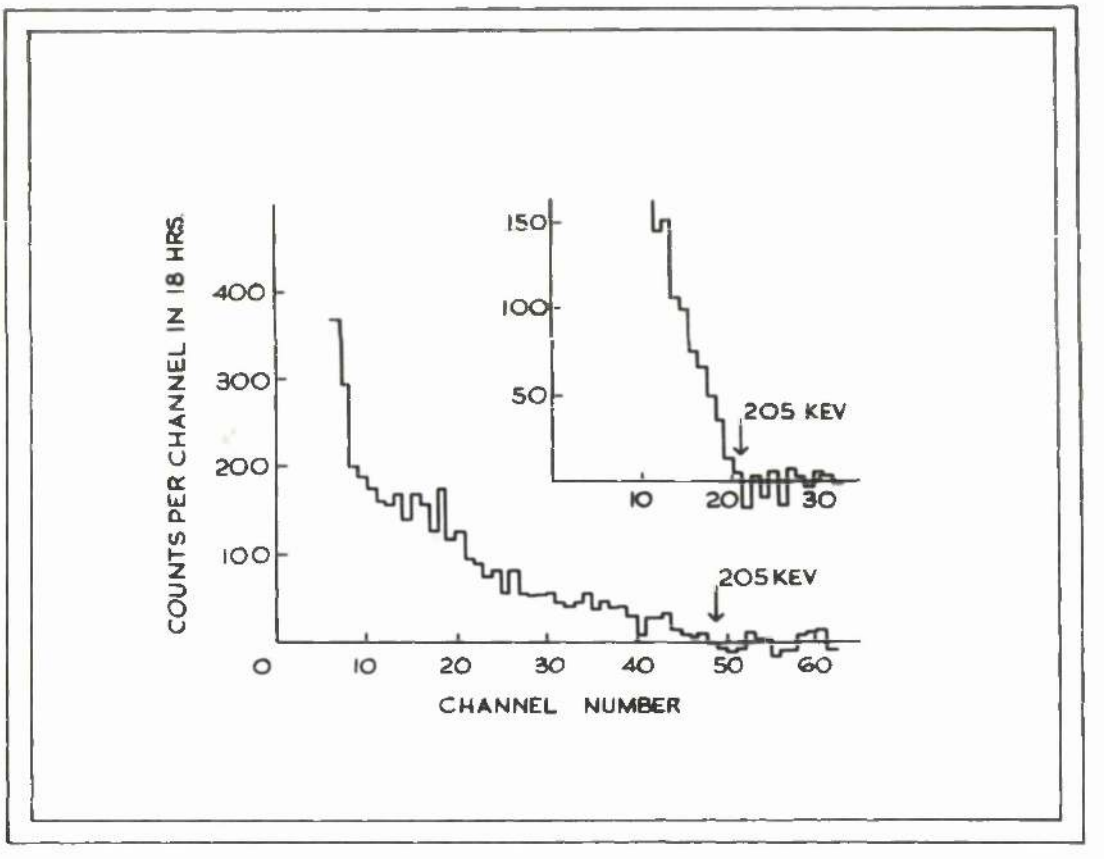

COUNTS PER CHANNEL IN 18 HRS
400
300
200
100
0
10
20
30
40
50
60
CHANNEL NUMBER
205 KEV
150
100
50
10
20
30
205 KEV

FIG 2.

The need for sources of very high chemical purity must be emphasised. Spectrographically pure La_2O_3 was found to give a spectrum showing several contamination peaks at energies <400 Kev, their total activity being considerably greater than that of the high energy gamma rays. An extremely pure sample, kindly supplied by Dr. W. Turchinetz of the University of Manitoba, Canada, showed practically no low energy contamination.

3. Beta-Ray Spectrum.

0.927 g of the pure finely divided La_2O_3 powder, suspended in amyl acetate containing a drop of Durofix as adhesive agent, was spread over 850 cm^2 of the removable copper lining of the proportional counter, the source being kept well within the effective counting volume. The average source thickness was 1.1 mg/cm^2. The counter was filled to a pressure of 4 atm., and calibrated by means of the 46.7 Kev γ-rays of Radium D.

The spectrum obtained (Fig. 2) indicated a maximum energy of (205 ± 10) Kev for the β-ray. The end point was observed, more clearly (Fig. 2 inset), by using a thicker source and lower amplifier gain. There was no indication of a β-ray of maximum energy ~ 1.0 Mev and partial half-life 1.2×10^{12} year, as reported by Mulholland and Kohman (1952), although such an activity would have been well within the limits of detection of the system used. A thinner source, 0.37 mg/cm^2, was used to examine the spectrum below 50 Kev. The shape must be distorted due to the necessary source thickness, but the results suggest/

/suggest that the spectrum exhibits a real rise at low energy. Further investigation with a sufficiently enriched source, as yet not available, is required to confirm that the spectrum shape is indeed similar to that of ^{87}Rb (3,4).

4. Coincidence Experiments.

The La_2O_3 source was placed between the two cyrstals. The output pulses from one photomultiplier, after amplification, were passed through pulse lengthener and brightener circuits and viewed on a cathode ray tube as a spectrum of bright spots in the X-direction. The pulses from the other multiplier were amplified and lengthened, and then applied in the Y-direction, so that a coincidence appeared as a bright spot whose displacement from the X and Y-axes was proportional to the energy spent in each crystal. The resulting pattern was filmed as a series of ten minute exposures and analysed.

For the investigation of possible coincidences between the 0.81 and 1.44 Mev γ-rays, only coincidences in which an energy >0.65 Mev was spent in both crystals were considered. The total coincidence rates for source + background and background respectively were 106 ± 10 and 96 ± 10 coincidences per 6 hours. From a knowledge of the geometry and the γ-ray counting rates in each crystal, a coincidence rate greater than twice the background rate would have been expected. Hence these two γ-rays cannot be in coincidence. Examination of the energy region 0.35 - 0.65 Mev gave no evidence of coincidences between/

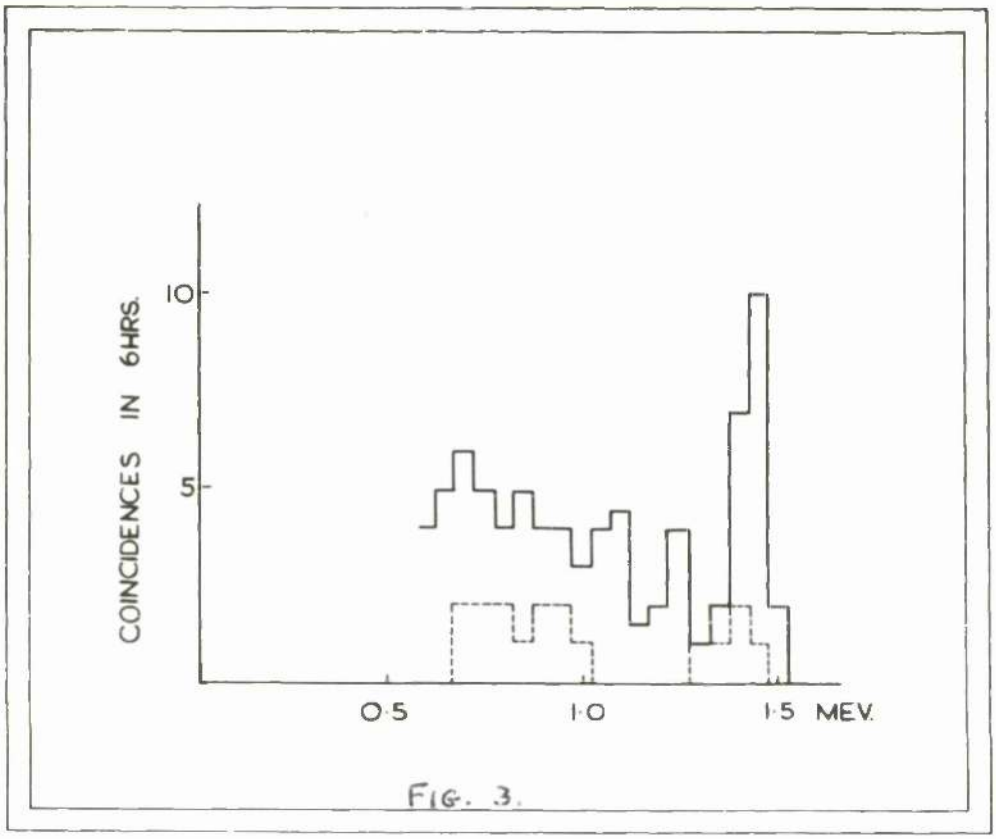

COINCIDENCES IN 6HRS.
10
5
0·5
1·0
1·5 MEV.

FIG. 3.

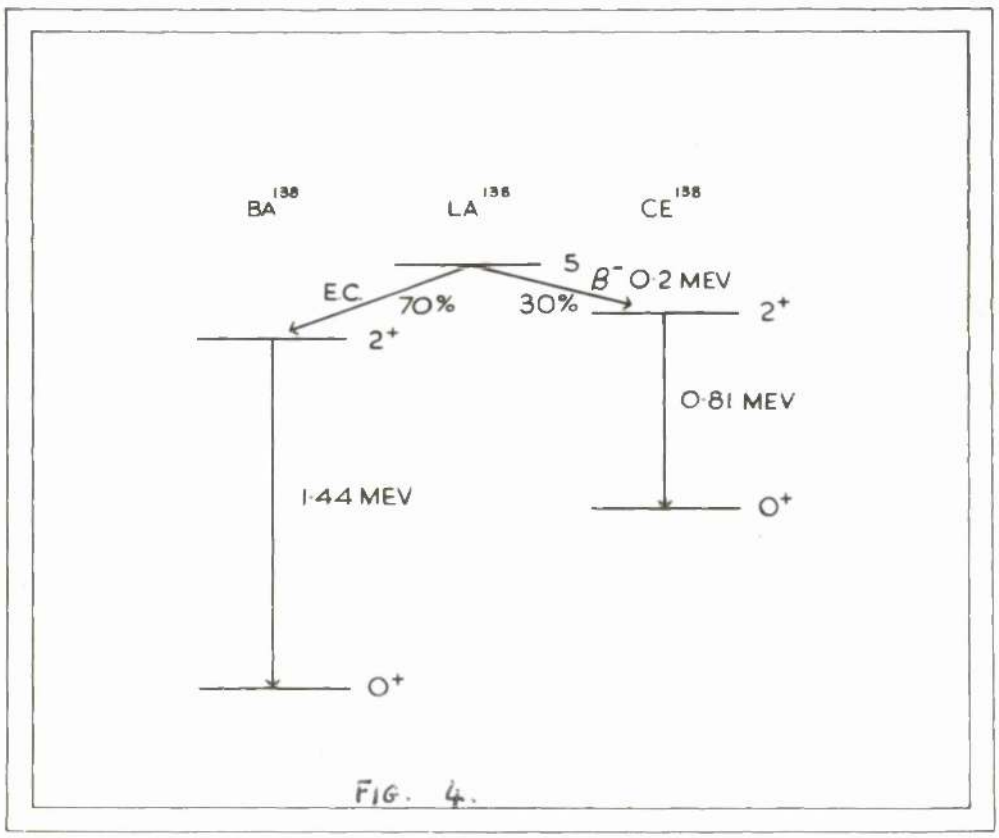

BA 138
LA 136
CE 138
5
β⁻ 0·2 MEV
EC
70%
30%
2⁺
2⁺
0·81 MEV
1·44 MEV
0⁺
0⁺

FIG. 4.

/between annihilation quanta, indicating that β^+-emission, if it occurs, must be of very low intensity, certainly < 1%, compared with electron capture. In the K-γ coincidence experiment a lead absorber, 3 mm thick, was placed between the γ-ray counter and the source to prevent any iodine escape x-rays being observed in the crystal which detected the x-rays since the energy of such x-rays is close to that of barium K x-rays. The spectrum of pulses in coincidence with the barium K x-rays (Fig. 3) clearly shows that only the 1.44 Mev γ-ray follows K-capture. The number of coincidences under the 1.44 Mev peak alone is 21 in 6 hours, compared with a background of 4.

To investigate β-γ coincidences, a relatively thin source would be required to permit detection of the β-rays. Because of the very restricted source area available in such an experiment, it was not considered to be feasible.

The decay scheme proposed on the basis of these experiments is shown in fig. 4.

5. Decay Constants.

Since the 1.44 Mev γ-ray follows electron capture, the specific activity of this branch can be simply determined by comparison with the ^{40}K 1.46 Mev γ-ray. Using the 2 inch NaI crystal, spectra were taken with sources of La_2O_3 and KCl under identical geometrical conditions. The detection efficiency of the crystal was assumed to be the same for each γ-ray. By measuring the total counts under the ^{138}La 1.44 Mev and ^{40}K/

/^{40}K peaks, knowing the mass of each source, and using a value of 3.47 gammas/sec/g K for the specific activity of potassium (mean value of the results of Backenstoss and Goebel;[5] and McNair et al[6]), the specific activity of the electron capture branch was found to be 30.9 ± 1.2 disintegrations/min/g La. Since the abundance of ^{138}La is 0.089%[7] the electron capture partial half-life is (1.64 ± 0.06) x 10^{11} years. This result is in agreement with that of 2 x 10^{11} years determined by Pringle[2] but not with the value of 4 x 10^{13} years estimated by Selig[8] from the observation of BaK x-rays using a proportional counter.

By comparison with the ^{40}K γ-ray spectrum extrapolated to zero energy the counts in the Compton distribution of the ^{138}La 1.44 Mev γ-ray were estimated. Hence, knowing the total counts N_1, N_2 due to the 1.44 and 0.81 Mev γ-rays respectively the partial half-life of the γ branch can be deduced from the formula

$$t_\gamma = \frac{N_1}{N_2} \cdot \frac{1 - \exp(-\mu_2 m)}{1 - \exp(-\mu_1 m)} \cdot t$$

where t and t_K are the partial half-lives of the β and K-capture branches and μ_1, μ_2 the total mass absorption coefficients for the 1.44 and 0.81 Mev γ-rays respectively; m is the average thickness in g/cm^2 of the NaI crystal. The partial half-life thus deduced was (3.5 ± 0.3) x 10^{11} years, the corresponding specific activity being (14.6 ± 1.2) disintegrations/min/g La.

Using the thinner source in the proportional counter, the β-rays with energy > 4 Kev were counter. By subtracting the BaK x-rays detected following electron capture, extrapolating the β counting rate to zero energy, and correcting for self-absorption and backscattering (45%) (6) a specific activity of (12.4 ± 2.1) betas/min/g La was obtained. The large probable error is mainly due to poor statistics since the observed counting rate from the source was only 2.5 c.p.m. compared with a background of 29.7 c.p.m. The partial half-life is (4.1 ± 0.7) x 10^{11} years. The specific activity is perhaps slightly lower than that found by the previous method. The suggested spectrum shape would provide a simple explanation, since even the thinner source would absorb an appreciable fraction of those β-rays with very low energy.

From these results, using the mean of the two values for the activity of the β branch, it is concluded that approximately 70% of the disintegrations proceed by electron capture to ^{138}Ba and 30% by β-decay to ^{138}Ce.

6. Discussion.

The spin of the ground state of ^{138}La has been reported to be 5 (9). The shell model predicts ($d_{3/2}$, $G_{7/2}$) configuration and hence even parity. Since the spin of the first excited state of an even-even nucleus is almost invariably 2+ (10) both the electron capture and β-transitions might be expected/

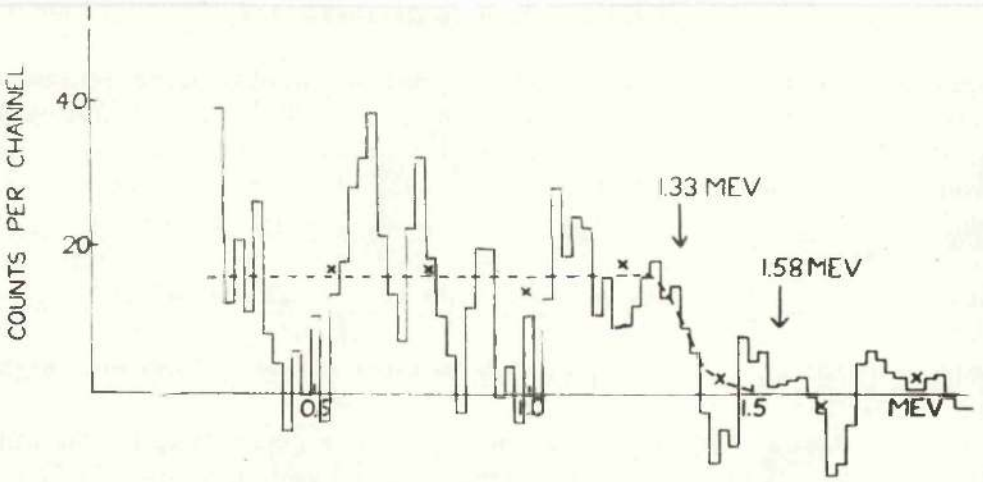
/expected to be $\Delta I=3$, no i.e. unique 2nd forbidden. However, the experimental log ft value for the β-decay is 19.0, corresponding to a 3rd forbidden transition, not 2nd forbidden. Goeppert-Mayer [11] has pointed out that the odd parity of the ground states of $^{122}_{51}Sb_{71}$ and $^{124}_{53}I_{71}$ indicates that the $h_{11/2}$ level can be occupied by an odd number of neutrons in the ground states of odd-odd nuclei. Hence in ^{138}La it is possible that the last odd neutron is in an $h_{11/2}$ level instead of $d_{3/2}$. The parity would then be odd, and a 3rd forbidden $\Delta I=3$, yes transition would result. Confirmation that the La β-spectrum has the same shape as that of ^{87}Rb would support this suggestion, since ^{87}Rb is the only known $\Delta I=3$, yes transition.

Since there are several excited states of ^{138}Ba <2.5 Mev above ground [12] the presence of only the 1.44 Mev γ-ray following electron capture indicates that β^+-emission to the first excited state of ^{138}Ba is energetically impossible. Decay to the ground state is obviously highly forbidden because of the large spin change ($\Delta I=5$) involved. The inability to detect annihilation quanta experimentally is thus easily understood. Similarly decay to the ground state of ^{138}Ce ($\Delta I=5$) with the emission of a 1.0 Mev β-ray is forbidden relative to the excited state. The absence of a 1.0 Mev β-ray (at a conservative estimate its intensity cannot be > 5% of the 0.2 Mev β-ray) is thus not unexpected.

Following the β-decay of ^{138}Cs, Langer et al [13] and Thulin [12] have detected a 1.44 Mev γ-ray, which on the basis of the systematics of Scharff-Goldhaber [10] was considered to be a transition from the first excited state of ^{138}Ba to the ground state. The present investigation supplies independent confirmation of this view. A 0.80 Mev γ-ray was observed by Handley and Olson [14] following β^+-decay of ^{138}Pr to ^{138}Ce. Thus the decay scheme proposed is in complete agreement with the most recent experimental data.

3.2 <u>A Search for Natural Radioactivity in Vanadium (Paper 1).</u>

A search for β-rays was carried out using the proportional counter situated in the underground site described in 3.1. The source of vanadium pentoxide was of average thickness 10 mg/cm^2. For the background a similar source of chromium oxide was used. The difference between source+background and background counting rates over a period of 8 hours was $+113 \pm 137$. Assuming a detectable limit of thrice the statistics and an isotopic abundance of 0.25% [15] then, taking into account β-ray absorption in the thick source, the half-life for emission of β-rays of energy > 50 Kev must be $> 2.4 \times 10^{14}$ years. In case the β-rays were emitted predominantly with very low energy as in the ^{87}Rb spectrum, the measurements were extended so that pulses of energy > 1.5 Kev could be detected, but no activity was observed.



The γ-ray spectrum of ^{50}V. Each cross denotes the average excess counting rate over a group of ten channels centred about its position.

FIG 5.

Using the 10 mg/cm^2 source the K x-ray region was closely examined, the difference between the source+background and background counting rates over 8 hours being +45±47. Assuming the same detectable limit, then, allowing for absorption in the source of the x-rays which would be produced in only 25% of the transitions, the K-capture half-life must be $> 4 \times 10^{13}$ years.

A search for γ-rays was undertaken with the scintillation spectrometer shielded by 4 inch lead lined with 4 inch steel. The 2 inch NaI crystal was surrounded by 110 g of vanadium pentoxide and by the same mass of chromium oxide for the background run. The source+background and background counting rates above an energy of 200 Kev were consistent over several periods adding up to 10 hours the totals being 28503±169 and 27644±166 respectively, an excess of 859±237. Since this excess counting rate was only 3% of the total background, the statistics were very poor as can be seen from the figure. There is no appreciable peak near 1.58 Mev but it seems significant that the spectrum drops sharply near 1.33 Mev, the energy at which the Compton edge would be expected. Also the result obtained from averaging the excess counting rate in each group of ten channels (Fig. 5) suggests that the counts below 1.33 Mev are spread evenly over the spectrum, as would be expected for a Compton distribution. Failure to detect β-activity eliminates the possibility of β-contamination. The α-activity of both the vanadium pentoxide and chromium oxide was found to be only 2 c.p.m., the minimum level of impurity we have found to be/

/be attainable. Although tests have been carried out on the oxides of six different elements which gave this α-rate of 2 c.p.m., no γ-activity has been found to be associated with the α-activity. By surrounding the crystal with a similar mass of potassium bicarbonate in the same geometry, the γ-activity was estimated to correspond to an electron capture half-life for ^{50}V of $(4.0 \pm 1.1) \times 10^{14}$ years.

Comparison of this half-life with the lower limit attainable in the proportional counter search for K x-rays shows the impossibility of detecting these x-rays using a thick source of unenriched material. It is estimated that if the search for x-rays was carried out using a minimum of 250 mg of vanadium pentoxide, 50 times enriched in ^{50}V, then a significant counting rate should be obtained.

3.3 The Half-Life of V-50. (Paper 2).

Since the completion of the preceeding work on V-50, the first excited state of Cr-50 has been discovered to be at 780 Kev. With the method described here, it is estimated that a factor of at least 100 improvement in sensitivity is attained and that any gamma-activity from V-50 with a half-life $< 2 \times 10^{17}$ years should be detectable.

1. Experimental.

The detector of the gamma-ray spectrometer is a Harshaw cylindrical sodium iodide crystal 9" in diameter and 6" high mounted in a 700 mgm/cm^2 electrolytic copper can and viewed by four E.M.I. type 9531B photomultiplier tubes.

When the detector was completely surrounded by 12 inches of mild steel inside the water shielded low background cell[16] at Aldermaston, the background between 660 KeV and 2.00 MeV was 720 c.p.m. The initial resolution obtained with the 660 KeV gamma-rays from Cs-137 was of the order of 10%, in good agreement with that normally expected for large crystals. By the end of the experiment this resolution had deteriorated to 12% as the output of the phototubes were no longer precisely matched.

As will be seen from the spectra, very good gain stability was attained with the crystal since the large volume of water shielding helps to maintain a constant temperature inside the low background cell. An energy drift of only 1% occurred in 300 hours continuous running.

The magnitude of the 1.46 MeV photopeak from K-40 shows that potassium has been almost completely removed from the crystal and the surrounding materials.

4998 grams of spectrographically pure vanadium metal nodules were packed into a thin perspex container designed to fit over the crystal to ensure reproducible source geometry and uniform thickness over the 1500 cm^2 available surface area. The spectrum in the range 450 KeV to 2 MeV was recorded in a 100 channel kicksorter in two separate runs, each of 64 hours duration.

Two 16 hour background spectra with the empty perspex container in positon were recorded over the same energy range.

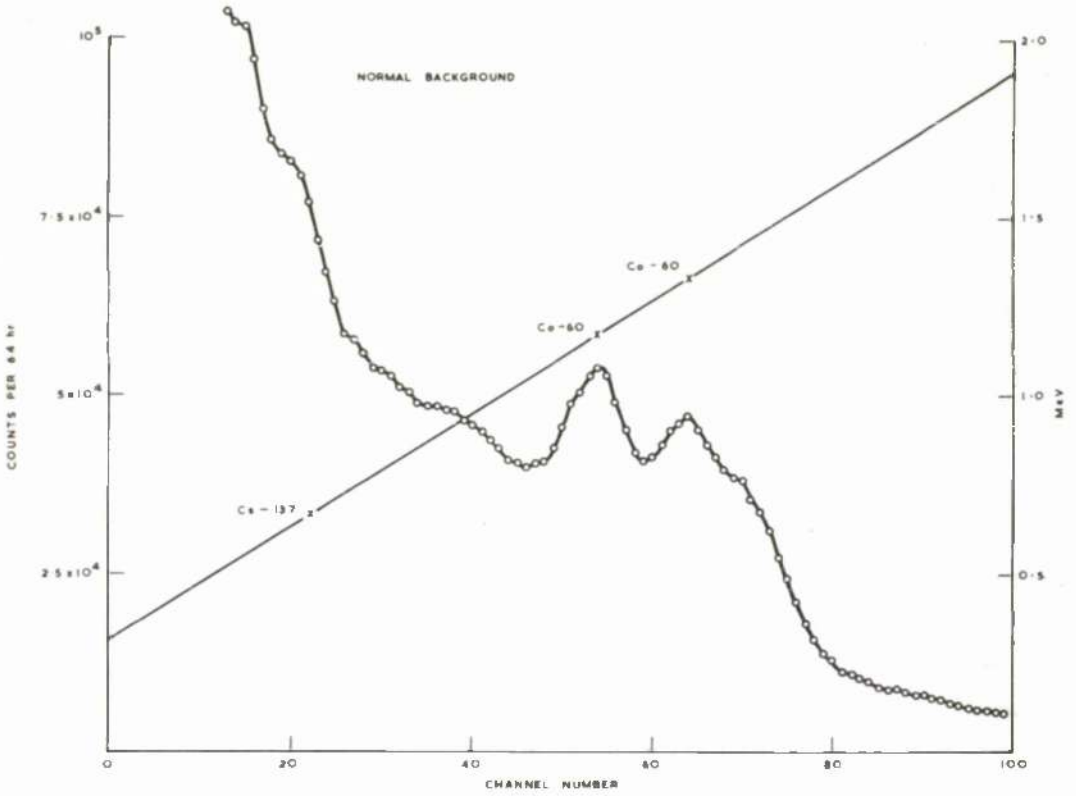


FIGURE 6

FIG. 6.

As large amounts of source material must introduce some additional degree of shielding and scattering effects to the normal background, a more realistic spectrum for comparison with the source is obtained by using an inactive material as a 'blank'. This must necessarily have a mass absorption coefficient for gamma-rays similar to that of the source, occupy the same geometry and have a high degree of purity. Copper filings prepared from electrolytic copper would be an ideal material in this case but copper oxide was used as this was more readily available. Consequently 5277 grams of metallurgical analytical reagent grade CuO was placed in the perspex container and the 'background' recorded in two separate 64 hour runs. These were alternated with the vanadium runs.

2. Results and Discussion.
Comparison of the CuO and the normal background spectra.

Fig. 6 shows the normal background spectrum obtained with the empty perspex container in position. As experienced by other low-background laboratories [17] with sufficiently sensitive equipment, low intensity gamma-peaks superimposed on the backgroun continuum are apparent. These are produced by the natural uranium and thorium series which occur to some extent in all common shielding materials. The 1.17 and 1.33 MeV gamma-rays arise from Co-60 on the steel bricks in the shield. Since this contamination is uniformly distributed over all the bricks it is assumed that it must have originated in the steel works where Co-60 is in fact used. However, the/

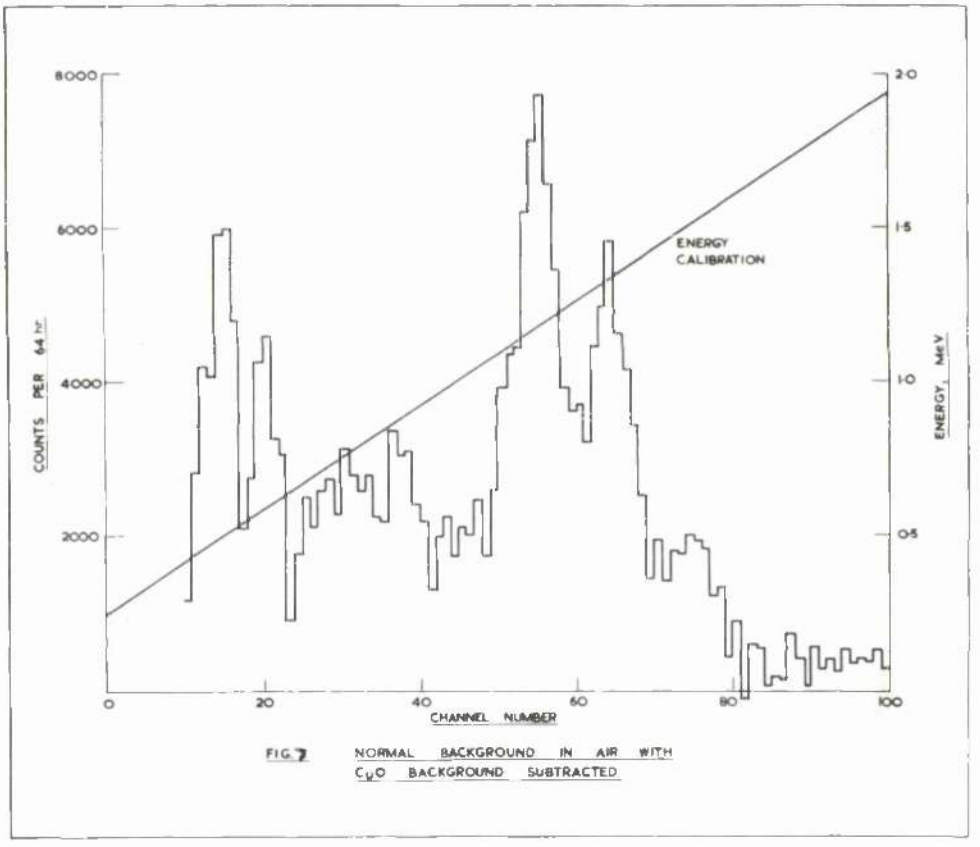


FIG. 7 NORMAL BACKGROUND IN AIR WITH C_4O BACKGROUND SUBTRACTED

FIG. 7.

/the overall reduction obtained in the background more than compensates for the disadvantage caused by the presence of the minute amount (10^{-10} gm. or one in 10^{17} parts of steel shielding) of Co-60.

When the spectrum obtained with the copper oxide is subtracted from the normal background (Fig. 7) the gamma-ray peaks stand out prominently and the background continuum is almost exactly compensated. This is to be expected for the cosmic radiation forms the continuum and localised contamination by natural radioactivity produces peaks in the spectrum.. When an additional absorber, such as the copper oxide, is placed round the detector, the background caused by the cosmic radiation is relatively unaffected as it is produced largely by high energy penetrating particles and showers created in the shielding by the interaction of fast particles. However, local gamma-rays are subjected to the normal attenuation and are considerably reduced in intensity. Hence, on subtraction of the spectra, definite peaks are created, due to the mass absorption differences. Alternatively, as is usual, had the normal background been subtracted from the copper oxide "source", negative peaks would be found corresponding to activity in the surroundings. Hence if a search was being made for a gamma-activity from a source corresponding in energy to any of the background peaks, a longer half-life than the true value would be obtained. These effects are usually negligible but become important in very low specific activity measurements. In the

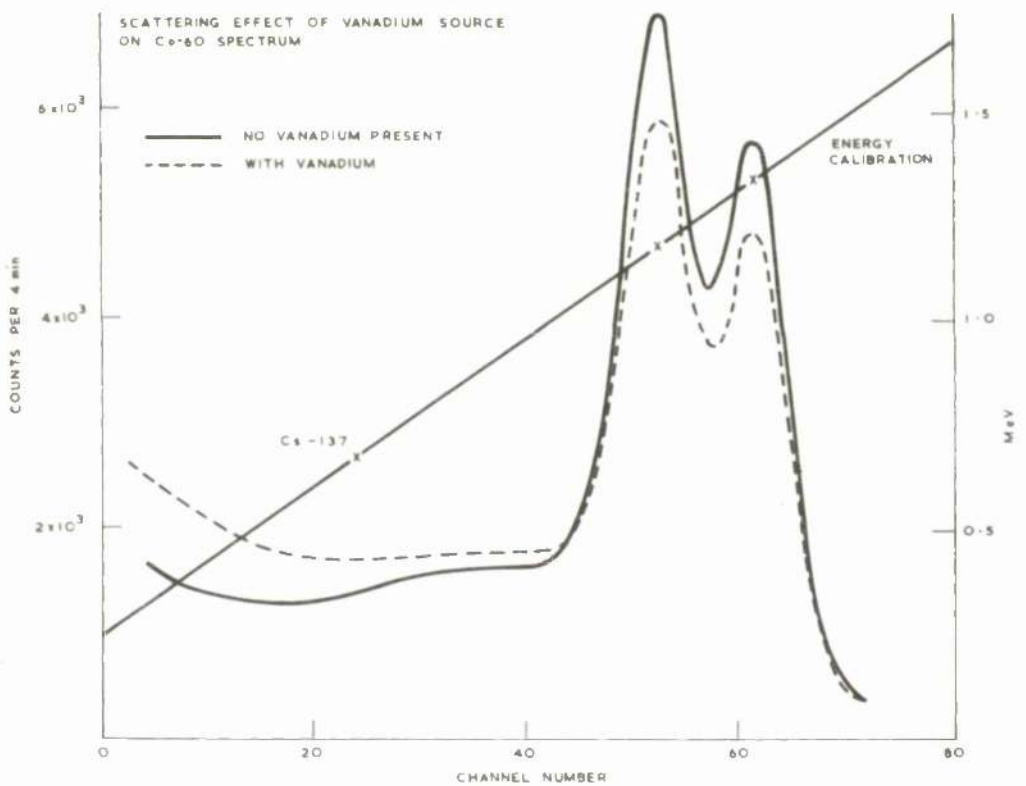


FIGURE 8

FIG. 8.

/the present case, the contribution is 2% of background which would involve about a 50% correction to the expected source activity. The method also provides a useful technique for determining whether or not gamma-activity arises in the crystal or in the shielding.

The effect of scattering of the background gamma activity by the source material must be considered in interpretation of the total count rates from the source. Fig. 8 shows the spectrum shape for an external Co-60 source with, and without, the absorbing material placed round the crystal. The low energy contribution increases with the absorber present giving a surplus count rate when the normal spectrum is subtracted.

To correct for the above effects when a very low specific activity source is being examined, a blank material with an identical mass absorption coefficient to the source must be used. If the background is overcompensated by the blank, spurious peaks will be produced and, if undercompensated, underestimation of the source counts may result. In practice it is difficult to exactly match the blank to the source as the materials available are usually of different chemical form but the difference in the average absorption can be found experimentally by using an external source of gamma-rays with an energy close to that of interest and normalising any gamma-rays occurring in the spectra on this basis. In the present experiment the Co-60 peaks were used for this purpose.

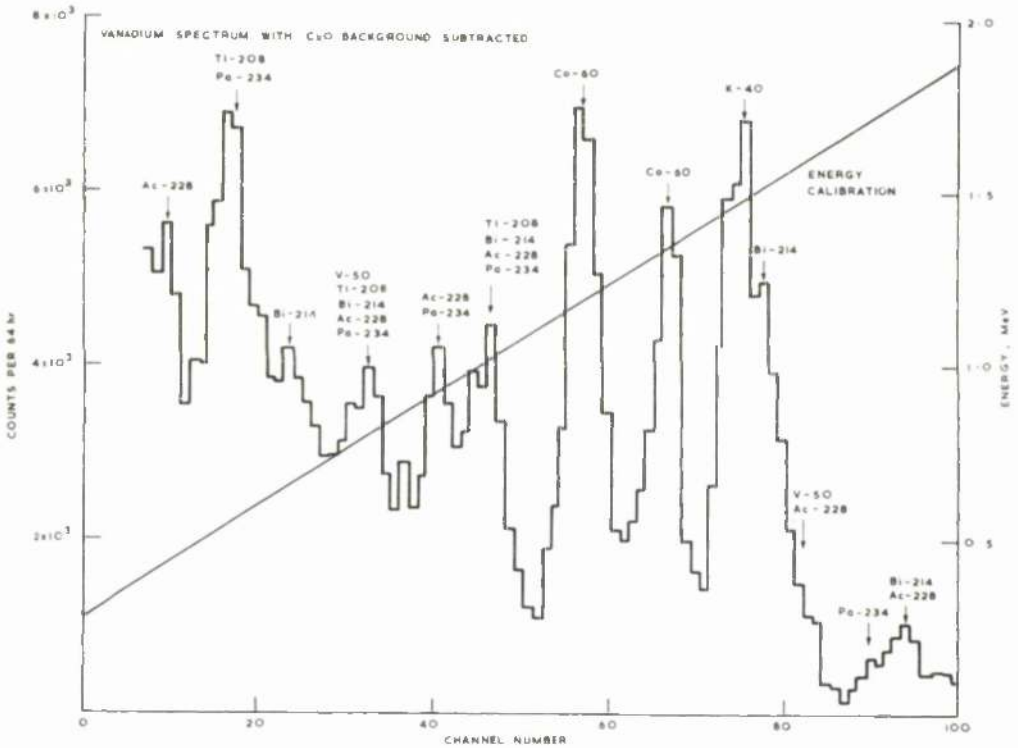


FIGURE 4

FIG. 9.

3. Vanadium Spectrum.

Several peaks (Fig. 9) were observed in the vanadium metal source after subtraction of the 'blank' background obtained with the copper oxide. The energies of the gamma-rays are consistent with those expected from the natural uranium (Pa-234, Bi-214) and thorium (Ac-228, Tl-208) series in addition to potassium-40. These impurties are found in most materials (18). Activity from a sample of vanadium pentoxide examined was five times greater than that in the specpure sample of vanadium metal.

The presence of the Co-60 peaks proved that the copper oxide is overcompensating the background consequently a correction factor must be applied. This latter factor was obtained by comparison with the intensity of the Co-60 peak remaining after subtraction of the copper background from the natural background. Allowing for the energy dependence of the absorption coefficient permitted correction of the remaining peaks for this background contribution. In the foregoing discussion it was assumed that the 'blank' material is very pure, which seemed likely as no negative peaks were observed.

After this correction, surplus counts still remained in the main peaks showing the vanadium itself contained the naturally radioactive impurities. Although the peaks are very prominent the total amount of uranium, thorium and potassium impurity are respectively less than one part in 7×10^7, 5×10^7 and 10^7 parts by weight of the vanadium and/

/and would not be clearly identifiable in previous equipment used for this type of study. This amount of contamination is at least an order of magnitude below the maximum quoted by the manufacturers of the spectrographically pure material.

Unfortunately the K-40 peak at 1.46 MeV overlaps the 1.59 MeV peak expected from vanadium. Interpretation is further complicated as thorium in equilibrium with its products has a 1.58 MeV gamma-ray and also uranium has an 0.76 MeV gamma-ray almost coinciding in energy with the first excited state of Cr-50. However, reasonable estimates of the contribution to these peaks from the impurity was made by the following procedures.

1. The potassium peak was subtracted on a resolution basis leaving the remaining counts in the 1.59 MeV photopeak.
2. Spectra of natural uranium and thorium samples in equilibrium with their daughter products were recorded under similar conditions used for the vanadium and the intensity of the 1.58 MeV and the 0.76 MeV gamma-rays relative to the 0.91 MeV and 1.00 MeV peaks were deduced. The contribution from the uranium and thorium to the 1.59 MeV and 0.78 MeV region were subtracted from the spectrum obtained with the vanadium source and any remaining counts attributed to gamma-ray activity from V-50.

Results for two separate spectra for V-50 combined with two different background runs corrected in the foregoing manner are quoted in Table I.

TABLE I.

	Corrected counts/64 hrs. in 0.780 MeV peak	Corrected counts/64 hrs. in 1.59 MeV peak
V_1-CuO(1)	2950	3664
V_1-CuO(2)	3991	3486
V_2-CuO(1)	4906	3402
V_2-CuO(2)	4515	2333
	mean 4090 ± 410	mean 3221 ± 275

There is still a positive residual count rate if the corrections are estimated with a view to minimising the tabulated results. It is considered most unlikely therefore that the corrections applied could be sufficiently in error to account for the excess number of counts. This leads to the conclusion that V-50 in radioactive and emits gamma-rays of 0.78 MeV and 1.59 Mev.

4. Crystal Efficiency.

Calculation of the gamma-disintegration rate from the source material requires a knowledge of the self-absorption, the geometry, and the crystal detection efficiency. These combined factors were measured experimentally by mixing a known aliquot of an absolutely standardised Cs-137 solution with the copper oxide used for the background run. A detection efficiency of 11.7% was derived for the 661 KeV/

/661 KeV photopeak. A higher energy calibration at 1.45 MeV was measured using potassium chloride. The total disintegration rate from the potassium source was calculated from the data given in Strominger's Table of Isotopes [19]. From the number of counts in the K-40 photopeak, the detection efficiency was calculated to be 5.5%. The overall efficiency for the 0.78 and 1.59 MeV gamma-rays was deduced from these two calibrations and the absorption coefficients to be 9.9% and 4.1% respectively.

5. Half-Life.

Using 0.25% for the abundance of V-50 in natural vanadium, the results of Table I yield a half-life of $9.6 \pm 1.6 \times 10^{15}$ years for electron capture decay to the 1.59 MeV level of Ti-50 and $1.8 \pm 0.4 \times 10^{16}$ years for the beta transition to the 0.78 MeV level in Cr-50.

If no corrections had been applied to these photopeaks, lower limits of 1.7×10^{15} and 1.4×10^{15} years respectively would have been obtained. Each of these limits is of the order of the results quoted for most previous experiments and it is probable that earlier activity attributed to vanadium is in fact caused by contamination with natural activity.

6. Conclusions.

Although the source was spectrographically pure, natural uranium, thorium and potassium were present in sufficiently large amounts to be serious. Had the vanadium been absolutely pure a half-life as long as 2×10^{17} years would have been/

/been detectable. As it is, after allowing for all the corrections, a definite surplus number of counts remain which can reasonably be attributed to the radioactivity of vanadium. These results yield a half-life for the electron capture transition to the 1.59 MeV level of Ti-50 of $9.6 \pm 1.6 \times 10^{15}$ years and a half-life of $1.8 \pm 0.4 \times 10^{16}$ years for beta decay to the 780 KeV level of Cr-50.

When these measured half-lives are combined with the available decay energy determined by Johnson, log ft values of 22.5 ± 0.5 and 23.1 ± 0.5 are derived for the respective electron-capture and beta transitions. This is consistent with the fourth forbiddenness inferred from the measured ground state spin of 6+ for V-50 and 2+ for the first excited state of Ti-50 and Cr-50. Since Ti-50 is on the closed neutron shell N = 28, the shell model can be expected to be valid and the transitions are unlikely to be hindered by nuclear deformation effects. These experimentally measured half-lives are therefore in good agreement with the theoretically predicted values.

In the measurement of very low specific activities careful measurement must be made of the background as subtraction of the normal background without a 'blank' source overcompensates and can lead to artificially long half-lives. Alternatively if the 'blank' material has a mass absorption greater than that of the source, unreal peaks (see Fig. 2) can be produced. The latter effect can be avoided by using/

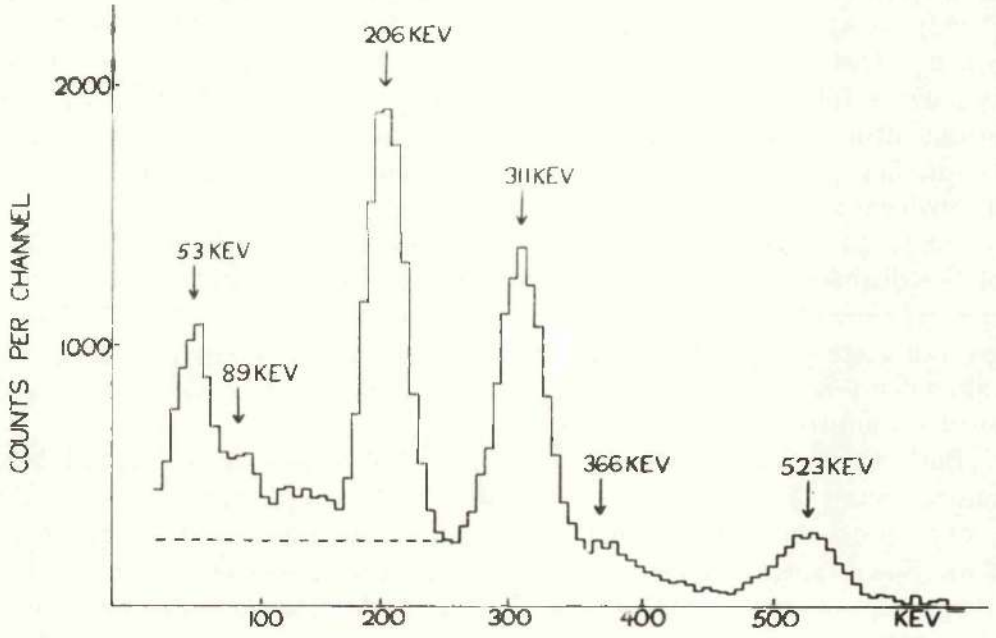
/using an external gamma source as a monitor and normalising the background to the source spectrum.

Contamination of the source by the natural radioactive series can best be checked by searching for alpha-activity as described earlier. As the gamma spectra from these series covers a wide range of energies it is necessary to account for the degree of its occurrence and effect in the investigations of low specific gamma-activities.

3.4 A Search for Electron Capture in Lu-176.

This experiment was undertaken to search for γ-rays or conversion electrons which could be attributed to the expected electron capture branch.

Both the scintillation and proportional counters described in section 3.1 were located in the underground laboratory at Glasgow. The γ-ray spectrum from 500 mgm. of lutecium oxide placed on top of the 2 inch NaI crystal is shown in the figure. The source counting rate was 700 cmp compared with the background of 80 cmp. The energies of the γ-rays were accurately determined to be 206±5 and 311±7 Kev in good agreement with the values of 203 and 306 Kev found by Arnold[20]. The peak at 523±10 Kev was due to simultaneous detection of these two γ-rays by the crystal. Similarly that at 366 Kev corresponded to addition of the 311 Kev γ-ray and a 55 Kev x-ray.



The γ-ray spectrum of ^{176}Lu.

FIG. 10.

The addition peak for the 206 Kev γ-ray and an x-ray would occur in the valley between the two γ-ray peaks and was obscured in the present spectrum. The incompletely resolved peak close to the K x-ray peak was due to 89 Kev γ-rays which were not internally converted. There was, therefore, no indication of any peak not attributable to the known γ-radiation.

120 mg of Lu_2O_3 was spread over an area of 800 cm^2 of the proportional counter lining giving a uniform source of thickness 0.15 mg/cm^2. The anticoincidence Geiger ring was not required. The pulses were fed along a 400 feet cable to the Hutchinson-Scarrott kicksorter. There was no indication of any electron peaks other than those known to be due to internal conversion of the 89 Kev γ-ray.

The half-life was estimated from the observed γ-ray spectrum. Allowance was made for the Compton distribution due to the 206 Kev γ-ray (Fig. 10). Then the remainder of the counts in the spectrum represent the total number of 311 Kev γ-rays detected by the crystal either alone or in coincidence with a 206 Kev γ-ray or an x-ray. A source of potassium chloride was placed in the same geometry and its spectrum taken. Knowing the specific γ-activity of potassium and calculating the average thickness of the crystal, the geometry was deduced using a value of μ_m, the mass absorption coefficient in NaI for 1.46 Mev γ-rays, interpolated from the tables given/

/given in the Appendix to Siegbahn[21]. Hence, using the value of μ_m for 311 Kev γ-rays obtained from the same tables, the half-life was calculated to be $(2.1\pm0.2) \times 10^{10}$ years. This is in excellent agreement with the value of $(2.15\pm0.10) \times 10^{10}$ years, found by Arnold[20] also from examination of the γ-ray spectrum, but not with that of $(4.6\pm0.3) \times 10^{10}$ years obtained by Dixon et al[1] from examination of the β-spectrum. The half-life was also calculated from our observed β-spectrum, but an -rate of 40 c.p.m. was obtained from the source. In the course of investigations on the natural radioelements with $Z<80$ it has been found necessary to check the purity of all sources, measurement of their α-activity, which is due to slight contamination by natural radioelements of the uranium and thorium series, being the most sensitive method. High α-rates have often been found to be associated with contamination, either β or γ, and hence the half-life calculated from the ^{176}Lu β-spectrum (2.8×10^{10} years) cannot be considered significant.

It is concluded that there is no evidence of γ-rays or conversion electrons which could be assocaited with an electron capture branch and that the branching ratio of $(3\pm1)\%$ proposed by Dixon et al[1] might rather be regarded as an upper limit. There appears to be a marked disagreement between the half-lives reported from γ and β-ray measurements. Unfortunately for the reason given above, it was not possible in the present investigation to determine the β-ray half-life.

Chapter 3.

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Chapter 4.

The Double Isobars.

4.1 Cd-In-113.

The main object of this experiment was to examine Cd-113 for possible β lability.

1. Experimental.

1.89 gm of 'Specpure' Cd O, obtained from Johnson Matthey & Co. Ltd., in the form of a very fine powder, was carefully spread over 1000 cm^2 of the removable copper lining of a large proportional counter, using a dilute solution of Amyl Acetate and Durofix as an adhesive agent. The counter was 14.4 cm in diameter and had 32.1 cm effective counting length between the field correcting tubes. A double ring of geiger counters operated in anti-coincidence was arranged aground the proportional counter, which was situated in the underground laboratory. The amplified pulseswere displayed on an oscilloscope and recorded on film. With the counter at a pressure of 40 cm. argon-methane mixture, the background above 1.2 KeV was 19.3 c.p.m. The K X-radiation of 9.25 KeV from a Ge-73 source was used for calibration purposes.

It has been found in the past that the presence of a stable source material on the copper lining changes the background spectrum in the following ways:

1. K and L X-radiation, characteristic of the source material, are excited by the cosmic radiation and/or any possible radioactive impurities in the source or counter construction materials.

2. The source material has an overall shielding effect caused by slight absorption of the cosmic ray background or absorption of any active contaminant present on the counter construction materials.

These difficulties can be overcome by the use of very pure materials and by using a stable material of similar weight and neighbouring atomic number to obtain a true background spectrum for comparison. In this experiment SnO_2 was used for the background.

The count rate recorded over 8 hours with the Cd O source was 8820 ± 156 and with the SnO_2 'blank' was 8731 ± 140. Hence the excess count rate due to Cd O is 11 ± 25 counts per hour.

Assuming Cd-113 is 12.26% abundant, that the saturation backscattering from copper would increase, the observed count rate by 50%, the geometry of the system is 2 pi and that we could distinguish a counting rate greater than twice the standard deviation of the mean we conclude that Cd-113 is stable towards beta-decay with a half-life.

$$> 1.3 \times 10^{15} \text{ years}$$

2.3 Discussion.

Recent work by Phillips and Hopkins [1] and by Bhatki et al [2] on the decay scheme of Sn-113 have established that the first excited state (spin and parity $\frac{1}{2}-$) of In-113 is 393 KeV above the ground state ($\frac{9}{2}+$). The transition is metastable with/

/with a half-life of 1.7 hours. Kjelberg et al[3] in a study of the decay scheme of Ag-113 have clarified the excited states of Cd-113. The first excited state ($\frac{11}{2}$) is 265 KeV above the ground state ($\frac{1}{2}$+).

It follows that if Cd-113 can undergo beta-decay with an energy greater than 393 KeV then a first forbidden transition would be possible. This would imply a log ft. value[18] of 7 ± 1 and hence a half life in the region of 10^{10} years. As this is well below the observable limit of 10^{15} years there cannot be sufficient energy to permit a beta-transition to the first excited state of In-113. Thus if beta-decay is possible it must be a fourth forbidden transition with log ft. ~ 23. For E ≤ 393 KeV this would imply a half-life > 5 x 10^{15} years. The experimental results support this deduction.

Alternatively, if In-113 undergoes K electron capture, allowance for the maximum error in the decay systematics would permit 100 KeV available energy for this mode of decay. Hence only a fourth forbidden transition could occur and the expected half-life will be > 10^{17} years - well above the sensitivity of the existing equipment. If L or M capture occurred the half-life would be even larger.

Examination of an Indium sample with a Be-windowed NaI crystal failed to reveal any gamma-rays in the range 20 to 300 KeV with τ < 1.4 x 10^{15} years.

4.2 In-Sn-115.

As there is some doubt about the value of the In-115 beta end-point energy a new measurement was undertaken and the half-life re-measured.

1. Half-Life Measurement.

0.9943 gm of finely powdered 'Specpure' In_2O_3 was spread uniformly over 900 cm^2 of the removable copper cathode of a proportional counter constructed from stainless steel. This counter was fitted with field tubes. It has a sensitive counting length of 45 cm. and is 8.2 cm. in diameter. The counter when filled to one atmosphere with argon-methane mixture had a background (using SnO_2 as a blank) of 27.0 ± 0.3 c.p.m. under an anti-coincidence assembly of geiger counters and 8" of steel shielding on all sides. The net count rate obtained with the In_2O_3 above 500 eV energy was 7.55 ± 0.50 c.p.m. above background. Assuming an isotopic abundance of 95.72%[4], 50% backscattering and a calculated source absorption correction[5] of 35%, the half-life is

$$6.0 \pm 0.4 \times 10^{14} \text{ years.}$$

This is in good agreement with Mattell's value.

2. Maximum Beta-energy.

An attempt to measure the end-point energy of the beta-spectrum was made with the source used for the half-life work but with the counting gas pressure at four atmospheres to increase the total energy spent in the counter and to reduce the wall effect. This showed that the end-point energy was/

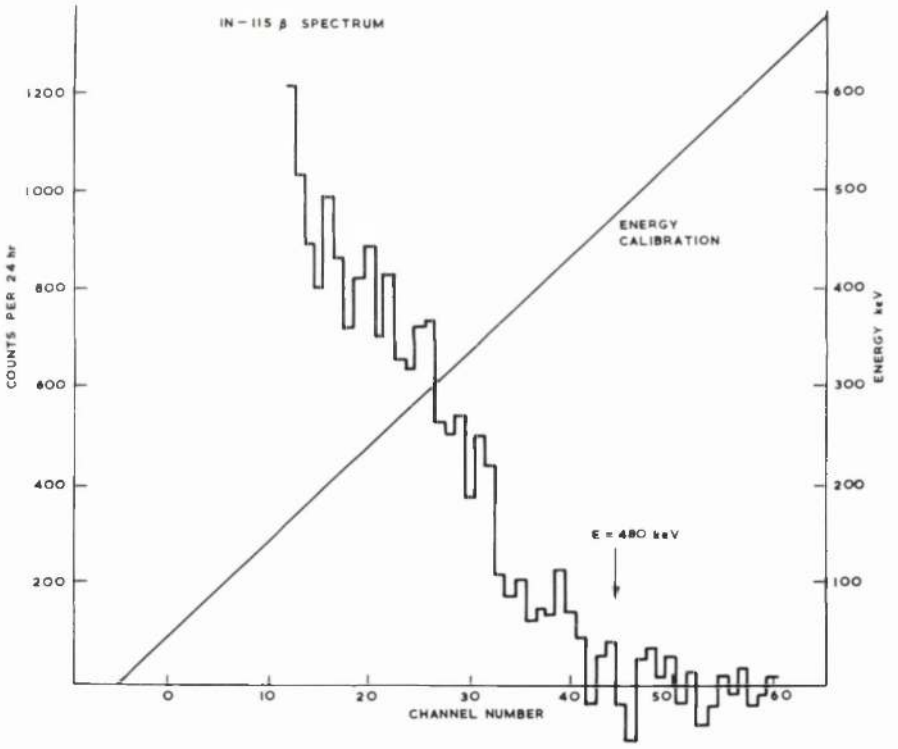
IN – 115 β SPECTRUM
ENERGY CALIBRATION
E = 480 keV
COUNTS PER 24 hr
ENERGY keV
CHANNEL NUMBER
1200
1000
800
600
400
200
600
500
400
300
200
100
0
10
20
30
40
50
60

FIGURE 1

/was greater than 300 KeV but the spectrum distortion was too large to permit an accurate measurement. It was decided therefore to use a plastic phosphor for the beta-energy determinations.

A plastic phosphor, type NE 102, 1" thick and 5" diameter was mounted on a 5" photomultiplier, type EMI 6364. The sides of the phosphor were coated with magnesium oxide reflecting paint and the top surface with an aluminium reflector 0.00003" thick. When shielded by 8" of steel inside the A.W.R.E. low background laboratory, a count rate of 230.7 ± 0.4 c/min 50 KeV was observed. An excess count rate of 10 cpm was detected when a source of 0.005" thick pure indium metal foil was placed on top of the aluminium reflector. The source and background spectra were each recorded for 24 hours with a 100 channel kicksorter. The difference spectrum is shown in Fig. 1. Energy calibration was effected with the Co-60 beta-ray end-point energy and the 440 KeV Compton edge from a Cs-137 source. From these results it is concluded that the In-115 beta-spectrum has a maximum energy of 480 ± 30 KeV.

A search for gamma rays > 30 KeV was made using a Be-window NaI crystal. It was concluded that In-115 is stable towards gamma-emission with a half-life > $3{,}1 \times 10^{16}$ years.

3. Discussion.

The ground state spins of In-115 and Sn-115 have been directly measured [6] to be 9/2+ and $\frac{1}{2}$+ respectively. Hence/

/Hence the In-115 decay to the ground state of Sn-115 is fourth forbidden. That the decay is direct to the ground state is confirmed by the absence of gamma-rays.

Assuming a half-life of 6×10^{14} years and a decay energy of 480 KeV, the log ft. is 22.7. This is consistent with the value expected for a fourth forbidden transition. The decay energy is in good agreement with that deduced from the work of Varma and Mandeville[7] on In-115m.

4.3 Sb-Te-123.

As no activity has been observed in either isotope it was decided therefore to search for possible beta, K capture and gamma activity.

1. Search for Beta-decay.

2.255 gm. of finely powdered 'Specpure' Sb_2O_3 was mounted on the cathode of the large proportional counter as used in the Cd-113 experiment. SnO_2 was used as the background source. No counts above 1.2 KeV were observed in 8 hours. Assuming twice the standard deviation of the mean would be detectable above the background, an isotopic abundance of 42.75%, 50% backscattering and 2 pi geometry, the upper limit for beta-radiation of energy < 50 KeV is

$$> 1.32 \times 10^{16} \text{ years.}$$

4.3 Search for K-capture.

The count rate obtained from a source of 2.26 gm. of natural TeO_2, containing 0.87% Te-123, mounted on the cathode/

/cathode of the proportional counter, filled to 40 cm. pressure with argon-methane mixture, was compared with that given by a SnO_2 coated cathode. An excess of 90 ± 25 counts per hour was obtained and found to be localised over the 28 KeV region of the spectrum corresponding to the K_α emission energy of Sb. Absorption of K X-rays in the source is only 1.4% and the detection efficiency of the counter was calculated to be 3.6%. Assuming a fluorescent yield of 85.5% and that all the Auger electrons would be detected by the counter, calculation yields a half-life for the K electron capture of Te-123 of $0.807 \pm 0.35 \times 10^{13}$ years where the error is the standard deviation.

Since the X-ray detection efficiency is very low for the above conditions, the experiment was repeated with the gas pressure at 4 atmospheres. This increases the efficiency to about 24%. Taking into consideration the corrections mentioned a value of $1.00 \pm 0.27 \times 10^{13}$ years was calculated.

As mentioned previously most of the 'Specpure' materials contain small amounts of alpha-activity. It was thought that this may be sufficient to excite the fluorescent X-rays to Te and account for the apparent K-capture activity. Consequently a source of 1.88 gm of Te metal powder was counted and Sb_2O_3 used for the background. The previous sources had similar alpha-activities of the order of 1 c.p.m. per gram. This Te source had an alpha-activity smaller than the Sb_2O_3, hence if the X-rays were excited by the alpha-/

/alpha-particles, a negative result would be expected. Once again excess counts of 75 ± 37 counts per hour were detected in the 28 KeV region of the spectrum giving a half-life of $1.64 \pm 0.49 \times 10^{13}$ years. No surplus counts were observed outside this energy range.

Since completion of the proportional counter experiment, a scintillation counter with a ¼" thick and 2" diameter NaI (Tl) crystal with a 0.005" beryllium window has been acquired. As this has a detection efficiency of almost 100% it was decided to repeat the search for K X-rays. This spectrometer was situated in the low background laboratory at AWRE and had a background of 89 c/hr. under the 28 KeV region. 0.807 gm. of TeO_2 was placed in a thin polythene bag, cut to fit closely over the beryllium window. A similar source of 0.957 gm. of Sb_2O_3 was used for the background. A positive result of 36.1 ± 3.25 c/hr. was again observed in the K X-ray region of the spectrum corresponding to a half-life for K capture of $1.32 \pm 0.12 \times 10^{13}$ years. The count rates above 40 KeV agreed within statistics.

The weighted mean of these four results indicates that Te-123 undergoes K electron capture with a half-life of

$$1.24 \pm 0.10 \times 10^{13} \text{ years.}$$

4.4 Search for Gamma-rays.

The existence of energy levels in Sb-123 and Te-123 at 161 and 154 KeV respectively have been established[8,9,10]. It was decided therefore to look for the gamma-rays which/

/which would arise if decay to these levels was energetically possible.

The beryllium windowed crystal was placed inside an 8" steel shield in the AWRE low background cell. This crystal has about 75% detection efficiency for 160 KeV gamma-rays and a background above 10 KeV of the order of 50 c/min.

Observation of the spectra from Sb_2O_3 and TeO_2 sources with a 100 channel kicksorter indicated no excess counts. It was concluded that Sb-123 has a minimum half-life for gamma-disintegration in the 10 KeV - 200 KeV region of 1.5×10^{15} years and that Te-123 is stable towards gamma-emission with a minimum half-life of 2.5×10^{13} years. Geometry, source absorption and detection efficiency corrections have been included in these calculations.

4.5 Discussion.

The above results indicate that Te-123 is active and decays by K electron capture to Sb-123 with a half-life of $1.24 \pm 0.10 \times 10^{13}$ years. Since the minimum gamma-ray half-life is appreciably longer than this, the transition must be mainly between the ground states of Sb-123 and Te-123 as these have measured spins and parities of $7/2^+$ and $\frac{1}{2}^+$ respectively, the electron capture transitions should be second forbidden with the characteristic log ft value ~ 13. When this value is combined with the measured half-life calculation shows that the decay energy available for the transition is only a few electron-volts. However, since the K_α X-ray is/

/is observed, the transition energy must be > 27 KeV which implies a log ft = 18. It appears, therefore, that the transition rate is hindered as could happen if there is configurational mixing of the states involved in the transition.

4.4 Re-Os-187.

Re-187 is known to be unstable with respect to beta emission. The energy of the transition is very low and probably < 8 Kev. It was thought that examination of an evaporated metallic rhenium source in the proportional counter would permit detection of the β-rays if the energy is greater than 150 eV.

1. Experimental.

An electrolytic copper cylinder formed the cathode of the proportional counter which was constructed from brass tubing of length 22.9 cm. and 8.9 cm. in diameter. Guard and field correction tubes mounted on ebonite insulators fitted into each end giving an effective counting surface area of 360 cm^2. A 0.001" diameter phosphor bronze wire was used as the anode.

When filled to one atmosphere pressure with argon plus 10% methane and operated inside the low background cell, the count rate above 150 eV was 136.0 ± 0.4 c/min. and in the range 150 eV to 2 KeV was only 19.1 ± 0.2 c/min. without any anti-coincidence arrangement.

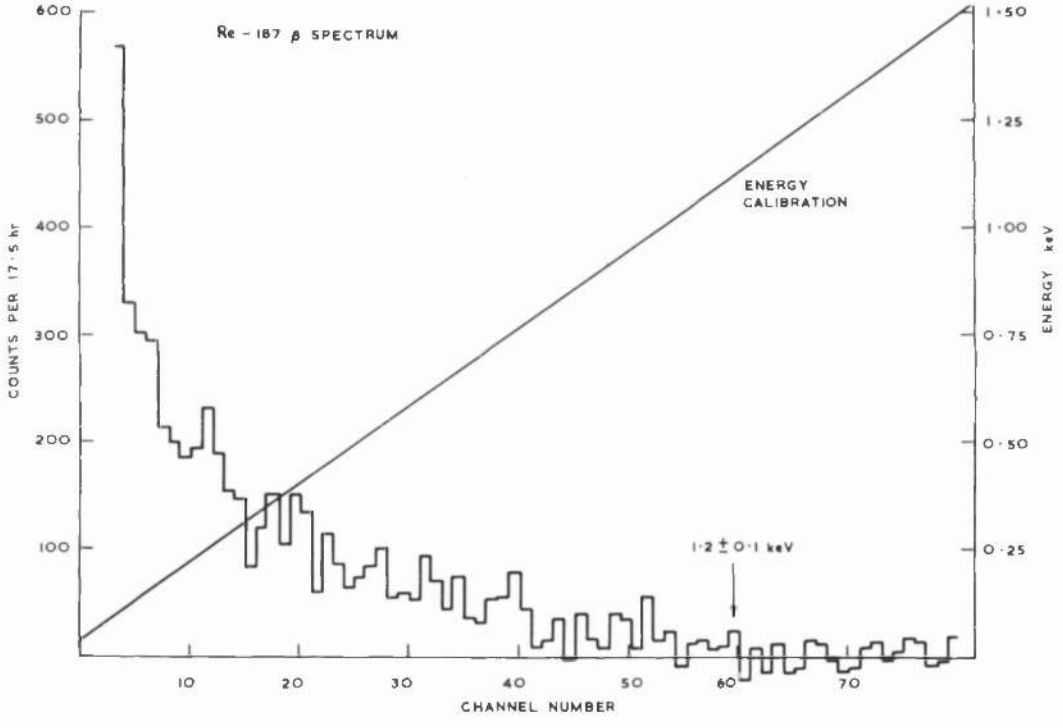
Re - 187 β SPECTRUM
ENERGY CALIBRATION
1·2 ± 0·1 keV
COUNTS PER 17·5 hr
ENERGY keV
CHANNEL NUMBER
600
500
400
300
200
100
1·50
1·25
1·00
0·75
0·50
0·25
10
20
30
40
50
60
70

FIGURE 2

Titanium K X-rays, excited by irradiation of a piece of titanium metal with the bremsstrahlung from a tritium source, were fired through a 0.010" beryllium window situated on the wall of the counter to give an energy calibration at 4.5 KeV. The 9.25 KeV X-rays from a Ge-73 electron capture source was also used. Resolution, full width at half height, was 20% and 15% for the respective peaks.

Background spectra were recorded on a 100 channel kick-sorter over the range 150 eV to 2 KeV for periods of 17.25 hours and 20.7 hours and in the range 1 KeV to 12 Kev for 6.8 hours.

A layer of approximately 40 gm/cm^2 thick spectrographically pure rhenium metal was vacuum evaporated onto the copper cathode of the counter which was then reassembled and the source spectrum recorded for an equal duration over the same energy range as the background. The spectra obtained with the source were alternated with the background runs, the source being cleaned off for the background and redeposited with rhenium for the beta search.

5.3 Results.

A consistent excess of 4.9 ± 0.1 c.p.m. observed above the background of 19.1 ± 0.3 c.p.m. in the low energy run is attributed to the beta-activity from Re-187. The spectrum with background subtracted is shown in Figure 2 where it can be seen that there is no count rate above background for energies > 1.2 ± 0.1 KeV. It is concluded that this is the/

/the maximum beta energy. The high energy run between 1 KeV and 12 KeV confirmed that no surplus counts occurred above 1.2 KeV. Owing to the very soft nature of the beta radiation, it was not considered possible to measure the specific activity of the source because of the serious self-absorption which must occur. However, a rough estimate of the half-life can be made by assuming the mean range for the average energy of the electrons deduced from a knowledge of the beta end point energy and that all the counts from this equivalent source thickness are detected. For an average energy of 400 eV and a range of the order of $0.5 \mu gm/cm^2$, obtained by extrapolation of the usual range-energy curves, the half-life is calculated to be of the order of 3×10^{10} years. The abundance of Re-187 is taken as 62.93% (4).

5.4 Discussion.

Since the spins and parities of the ground states of Re-187 and Os-187 are 5/2+ and 1/2- respectively, the beta decay should be first forbidden unique with a log ft. value in the range 8.5 ± 0.5 if characteristic of this group. The measured decay energy of 1.2 KeV and estimated half-life of 3×10^{10} years give a log ft. ~ 10 which is in fair agreement with the theory considering the uncertainty in the half-life and the doubt about the magnitude of the coulomb to the ft. value for such low decay energies. A half-life of 5×10^9 years would give a better fit with the theory and could be more accurately determined by counting rhenium in the vapour phase to avoid absorption losses.

Chapter 4.

References.

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Chapter 5.

Alpha Radioactivity Among the Medium-Heavy Elements.

5.1 Introduction.

The discovery by Hevesey and Paul in 1932[1] of the naturally occurring α-activity in Samarium and the subsequent discovery of pleochroic haloes in rocks which had contained Pm-145, prompted much discussion[2-6] on the possible alpha instability of elements below Lead ($Z = 82$). The conclusion reached was that most of the medium heavy elements in the periodic table are, on the average, unstable towards alpha emission but would be beyond the limits of detection by existing techniques since the half lives are expected to be in the region of 10^{16} years or greater. Fluctuations about this average could explain the alpha lability of Samarium and possibly a few of the missing β-stable nuclides.

Kohman[7] (1949) has deduced the approximate energy released by alpha disintegration by estimating the individual alpha particle binding energies from the empirical mass equation of Bohr and Wheeler[8]. He concluded that most nuclides above mass A = 140 are energetically capable of alpha-emission and are stabilised by the nuclear potential barrier resulting from the high charge. On the neutron deficient side of the β -stability region, α-activity must be an important mode of decay and may complete or replace electron capture or β^+ decay.

Detection and measurement of the decay rates and disintegration energies of α-emitters in the rare earth region are of interest since they provide an accurate measure of the/

/the binding energies and mass defects in a region considerably removed from the heavy element alpha emitter. Also the correlation of alpha decay rates and decay energies among the rate earths, assuming the theoretical decay rate formula[9], permits calculation of the "effective" nuclear radius for alpha particles for the daughter nuclei. However, longer half-lives than expected would not be uncommon in view of the occurrence of 'hindrance factors' for odd A nuclei[10]. Indeed, odd A nuclei usually have lower decay rates by $\sim 10^3$ than even A nuclei.

With a knowledge of the alpha disintegration energies of many of the heavy isotopes and the β-decay of a few it has been possible to calculate the relative masses of nuclides of given radioactive families. Unfortunately, in the medium heavy element region this information is not available to permit calculation of masses by closed decay cycles.

In an effort to obtain more experimental information Thomson et al (1949)[11] embarked on a programme of producing neutron deficient isotopes by deuteron bombardment of targets of the medium-heavy elements in the 184 inch cyclotron at Berkeley. They succeeded in separating and detecting alpha activities in gold, mercury, gadolinium and dysprosium. These activities were all of short half-life. Since the alpha to electron capture or β^+ branching ratio is very much smaller for elements in the gold region than for those in the rare earths (where α/E.C. $\sim$ 1%), the authors concluded that in the rare earths the alpha decay is due to amore moderate neutron deficiency which gives rise to/

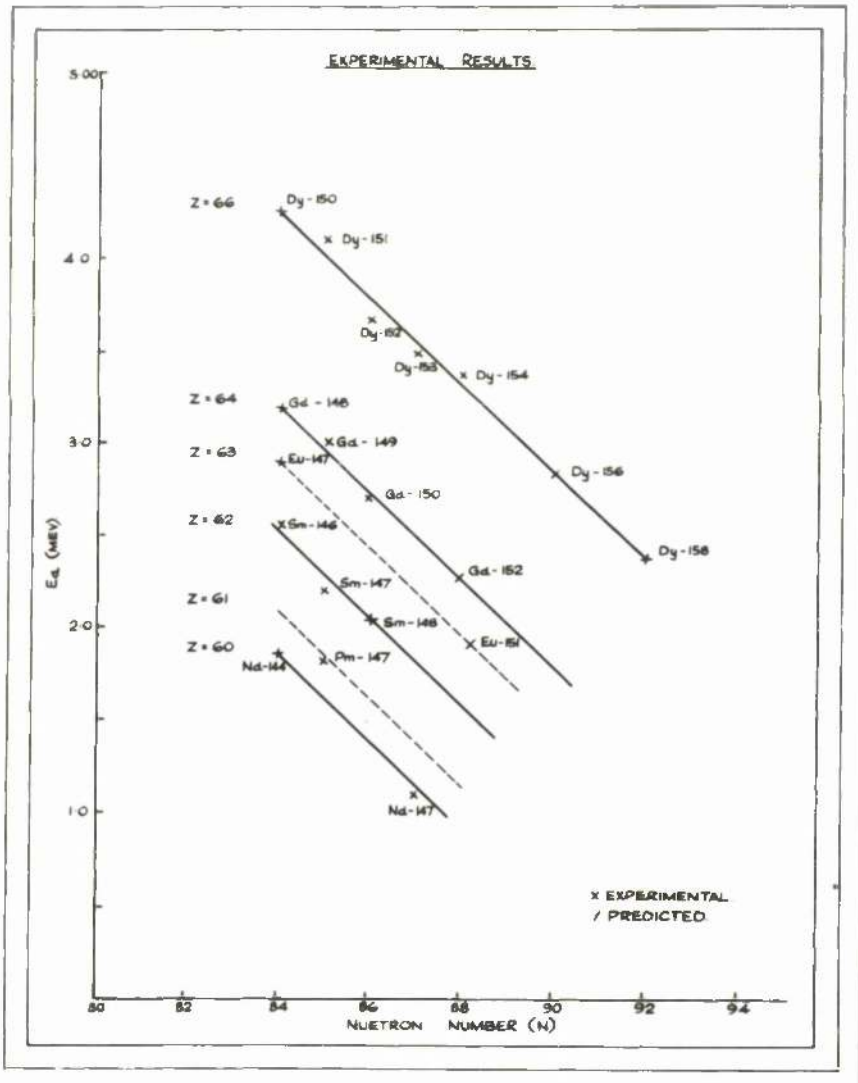

EXPERIMENTAL RESULTS
5.00
4.0
3.0
2.0
1.0
Eq (MEV)
Z = 66
Z = 64
Z = 63
Z = 62
Z = 61
Z = 60
Dy - 150
Dy - 151
Dy - 152
Dy - 153
Dy - 154
Dy - 156
Dy - 158
Gd - 148
Gd - 149
Gd - 150
Gd - 152
Eu - 147
Eu - 151
Sm - 146
Sm - 147
Sm - 148
Pm - 147
Nd - 144
Nd - 147
x EXPERIMENTAL
/ PREDICTED
80
82
84
86
88
90
92
94
NUETRON NUMBER (N)

FIG. 1.

/to longer competing electron-capture half-lives combined with exceptionally high alpha energy and hence a correspondingly shorter alpha half-life since the alpha decay probability is a very sensitive function of energy. This latter reason can be correlated with the stable configuration of 82 neutrons in such a way as to acquire the necessary disintegration energy in analogy with the isotopes which have extra high energy in the range 127-130 neutrons just above Pb at the closed shell of 126 neutrons, e.g. $Po^{211, 212, 213}$.

On this basis Thomson concludes the natural alpha-activity of Samarium arises from the isotope Sm^{147} with 84 neutrons.

From these and later measurements of Thomson, Rasmussen[9] and others on the alpha energies of the artificially produced alpha emitters in the medium-heavy region, it is possible to deduce the alpha decay energy available for the naturally occurring neutron-deficient isotopes by plotting alpha energy against neutron number for each element. This has been carried out for some of the rare earths (Fig. 1). Note that the maximum α-decay energy occurs just above the closed 82 neutron shell where the binding energy is lowest. Also, the trend of parallel lines for alpha decay energy to increase with decreasing neutron number is as predicted by the Fermi-Weizacker semi-empirical mass formula although the predicted energies are wrong since this formula does not allow for structural effects such as those ascribed to closed shells. It would appear, therefore, that the large alpha decay energy is undoubtedly a consequence/

/consequence of the decreased neutron binding energy just beyond the closed shell of 82 neutrons in analogy with the maximum at N = 128 just above the 126 neutron closed shell in the heavy element alpha-emitters[12].

Semi-theoretical information on the energetics of nuclei can be much more accurately obtained from the more recently developed empirical mass equation of Levy[13] 1957. This is fully discussed in Chapter 1, Section 1.2. The mass defect in terms of Z and A is given by:

$$\Delta M(A,Z) = a_0 + a_1 A + a_2 Z + a_3 A.Z + a_4 Z^2 + a_5 A^2 + \delta$$

where there is one set of coefficients for each shell region and δ depends on whether the nuclide is odd-odd, odd-even, even-odd or even-even. This formula reproduces the experimental results to within 0.5 MeV for 95% of the cases. Table I is a list of all the alpha unstable naturally occurring nuclides with their respective decay energies for Z<82 as predicted by this equation. The approximate half-life can be calculated from the simple formula of Condon and Gurney[14] or Gamow[15] using the value of the decay energy including the coulomb correction and allowing for the recoiling nucleus. It is the purpose of this work to examine some of these isotopes with a view to experimentally determining the disintegration energy and the half-life. For this study a proportional counter with a large area cathode was used. The merits of the various possible techniques were considered before deciding upon this form of detection.

5.2 Methods of Detection of Low Specific Activity a-Emitters.

As very low specific activities < 0.02 counts per second per gram are expected for the medium-heavy element alpha emitters the following requirements should be considered before deciding upon the most suitable technique.

1. Large amounts of source material are required to provide a sufficient counting rate for reasonable statistical accuracy.
2. Thin sources are necessary to prevent serious source absorption.
3. A low natural background is essential.
4. Long term stability of equipment is important to permit sufficiently long counting periods to accumulate the necessary statistics.
5. High purity samples are mandatory.

Nuclear emulsion methods are well suited to this type of study. The plates can be loaded with ~ 30 m.g.m. quantities of sample which can be left for several months before analysing. The nature of the activity is determined by the density of ionisation along the path and the energy by the track length. However, there are difficulties with this technique because fading of the tracks occurs with time, restricting the exposures to a limit of about three months. The stopping power of the plates alters with the degree of loading and shrinkage occurs with time hence energy calibration is difficult and necessitates the presence of a known alpha energy for calibration with a consequent rise in the effective background. As there is a/

/a three dimensional effect in recording, a total count determination is complicated and there is a limit to the shortest projected track length observable increasing the possibility of losing counts.

Ionisation chambers have also been used for this type of measurement. Once again, however, the source quantity is restricted as the source must be thin enough to prevent serious self-absorption, i.e. to the order of 100 μ gm/cm^2. For a possible 1000 cm^2 source area, 100 m.g.m. of source can be easily mounted. The method is not appreciably more sensitive than photographic emulsions since the geometry is only 2π although the disadvantages of energy calibration and fading experienced with photographic plates do not arise. The pulse height has been shown to be strictly proportional to energy and long counting runs can be made over a period of months if need be, although gas poisoning may occur and frequent refilling of the chamber is advisable to stop drift. Electronic pulse height recording is a big advantage over the tedious microscopic analysis required for photographic techniques.

A 4π gas filled proportional counter has been used[16] to measure the half-life of Sm^{147}. Here the practical limitations of producing large area thin films arise. The method has no outstanding advantages over previous methods and because of the source area restrictions, is almost certainly inapplicable to isotopes with half-lives several orders of magnitude greater/

/greater than Sm-147.

Kohman in 1953[17] suggested the use of a large proportional counter for this type of work but to date the method does not appear to have been used. The cylindrical proportional counter can have a very large cathode area and yet be a practical size. A cathode area of 5,000 cm^2 is possible which would permit gram quantities of source to be used. Since the pulse height is proportional to the energy spent in the counter and the counter has long term stability, a factor of about 30 times more sensitivity is obtained over the photographic emulsion technique and 5 to 10 times improvement over other ionisation chamber methods. This would permit observation of half-lives one to two orders of magnitude longer than observable with existing methods. Consequently a counter, which would be suitable for long-lived alpha half-life studies, was designed and is described below.

A table of results (Table II) and the measurement techniques used has been formed using literature published before June, 1961.

5.3 The Alpha Half-Life of Sm-147.

As can be seen from Table II, there is still some doubt about the value of the Sm-147 half-life for alpha emission. Since this decay rate is several orders of magnitude faster than those anticipated for the other isotopes listed in TableI, it provides a suitable means of testing the proportional counting equipment. Consequently a new determination of the Sm-147 half-life was undertaken using a gridded ion chamber and/

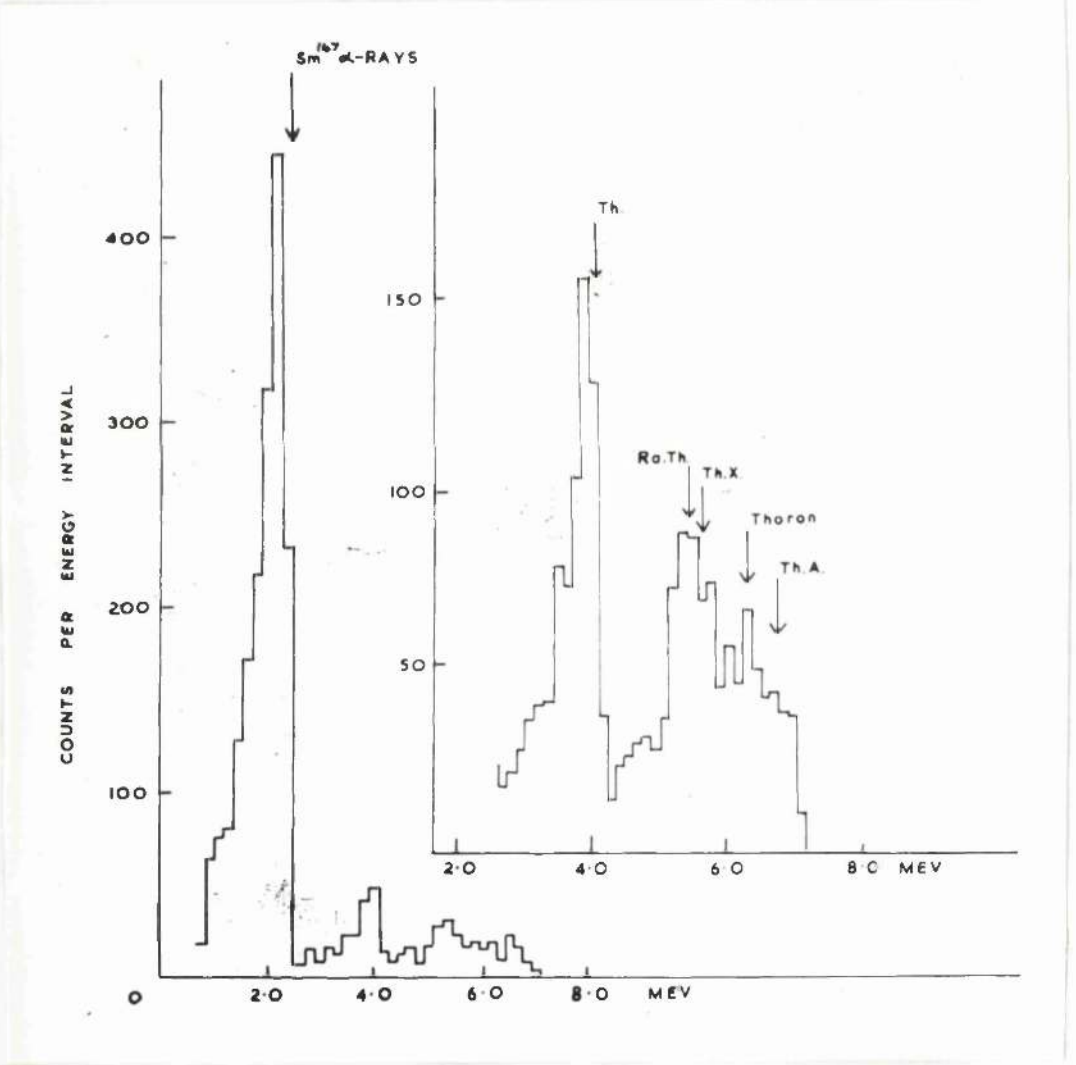

Sm147 α-RAYS
Th
Ra.Th
Th.X
Thoron
Th.A
COUNTS PER ENERGY INTERVAL
400
300
200
100
0
150
100
50
2·0
4·0
6·0
8·0
MEV

FIG. 2.

/and the result compared with that obtained by proportional counting.

1. Ion Chamber Method.

0.0108 gm. of spectrographically pure samarium oxide (Sm_2O_3), in the form of a very fine powder was weighed into a clean beaker to which was added a few cc. of amyl acetate mixed with a small drop of Durafix to act as an adhesive agent. This was carefully transferred to the cathode of the ion chamber and spread evenly over 80 cm^2 surface area to give an average source thickness of 135 $\mu gm/cm^2$.

The pulses from the chamber were amplified by a type 1430 head and main amplifier and fed into a 100 channel pulse height analyser. In addition to the Sm-147 peak at 2.20 MeV, peaks at higher energy corresponding to the natural uranium thorium and actinium series were observed (Fig. 2). Fortunately, the "tail" from these peaks flattens out in the 2.00 MeV region and an extrapolated correction (included in Table III) is easily applied. This activity is caused by impurity in the samarium and not produced in the source preparation as verified by doing a blank run. However, the impurity serves to increase the effective background and makes use of the normal background correction quite meaningless.

Spectra were recorded for several separate runs and repeated for different source thicknesses to check if there were any absorption losses. It is concluded from these results and from the absorption experiment to be described that losses/

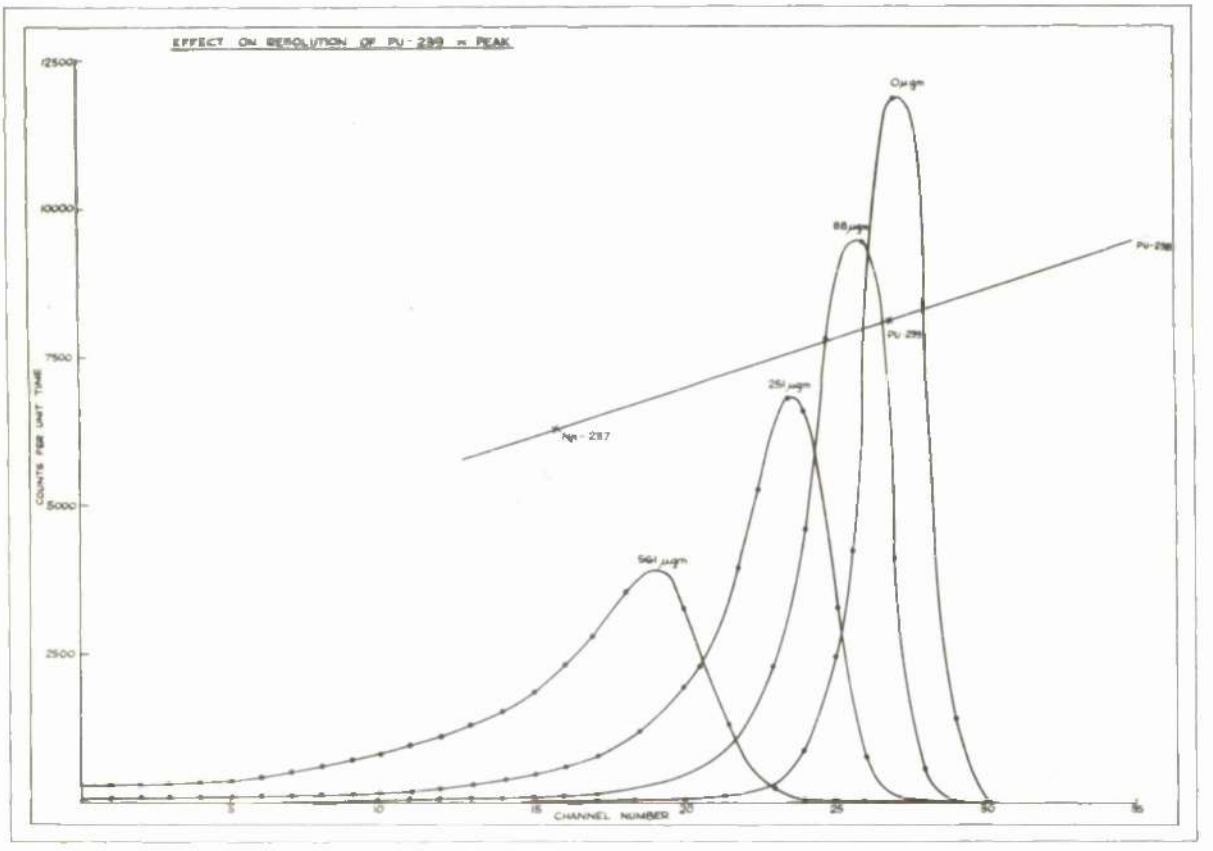

EFFECT ON RESOLUTION OF PU-239 α PEAK
12500
10000
7500
5000
2500
COUNTS PER UNIT TIME
0 μgm
88 μgm
251 μgm
561 μgm
Pu-238
Pu-239
Np-237
5
10
15
20
25
30
35
CHANNEL NUMBER

FIG. 3

/losses are less than 2%.

2. Calibration and Results.

Normally very thin sources of mixed Pu-239, 238 and 236 isotopes, electrodeposited onto a stainless steel disc, are used for energy calibrations and a linearity check of the chamber. The work of Karras et al[22] in which α-particles with energies of 1.797 and 1.474 MeV were produced by the B(n, α) reaction, verified linearity from 1.47 to 5.7 MeV.

When thick sources, ~100 μ gm/cm^2 are used, degradation of the spectrum results since there is a loss of energy by the α-particles traversing the source. The peak therefore falls in energy and the "tail" is incrased. Hence in energy calibration with thin sources this effect must be allowed for. It is also important to know whether the increased spread in the peak caused by absorption, is sufficient to result in losses from the total source count rate. This was investigated as follows:

An electrodeposited source of Pu-239, which had been absolutely calibrated in a low geometry counter, was placed in the chamber and counted for a fixed time to give good statistical accuracy. The resolution is about 1% full width at half-height for 5.00 MeV α-particles when filled to two atmospheres pressure with argon and 10% methane. Successive layers of gold were then evaporated over this source and the spectrum recorded for the same time between the respective layers. The gold thickness was measured spectrophotometrically using a monitoring VYNS-/

/VYNS-coated copper anulus placed beside the steel disc. Fig.3 shows the results.

For gold thickness up to 560 μgm/cm^2 the area under the peaks remains the same within statistics, if the "tails" are extrapolated to zero energy. It is concluded therefore that there should be no absorption losses in the samarium experiment when the data is interpreted in this way. Also the chamber geometry is measured to be 49% by comparison with the standard source.

From these results and the calculations of Table III the half-life of Sm-147 is found to be $1.19 \pm 0.04 \times 10^{11}$ years.

As there is a considerable energy shift in the peak with the addition of the gold layers it is clear that a correction must be applied to any energy measurement made on the basis of the electrodeposited source calibration. This is difficult to calculate accurately because of the difference in absorption of gold and samarium oxide and also because in the latter case, the activity is uniformly distributed throughout the source. Consequently a small amount of Pu-236, chosen to give approximately the same count rate as the samarium, was mixed with the samarium oxide during the source preparation. Since this must suffer similar absorption losses an accurate energy calibration was possible.

It is deduced that the energy of the Sm-147 α radiation is 2.20 ± 0.05 MeV.

3. Proportional Counter Method.

Since most of the suspected α-unstable nuclides are expected to have half-lives several orders of magnitude longer than Sm-147, a proportional counter was constructed with a 5,000 cm^2 surface area. This permits source amounts of about two grams to be mounted without introducing serious absorption losses thus giving workable count rates, e.g. 2 gm. of Nd_2O_3 with an assumed half-life of 5×10^{15} years should give a count rate of 13 counts per hour from the Nd-144 isotope. For equal counting periods, the increase in sensitivity over a nuclear emulsion method is a factor of about 50 although the latter method has some advantage since it does not require electronic equipment and there is little source absorption. However, since the background rate for both methods is similar this means the result can be obtained to the same accuracy by proportional counting in one fiftieth of the exposure time required for photographic methods.

Description of Counter.

The counter is constructed from brass tubing 19 cm. in diameter and one metre total length with a sensitive counting length of 84 cm. It is fitted with guard and field correction tubes, one of which is supported in a metal mounting fixed to the wall of the counter to permit removal of the end flange without breaking the 0.003" anode wire. This allows a 0.015" thick electrolytic copper cathode liner, which push-fits round the inner wall of the counter, to be easily removed when desired.

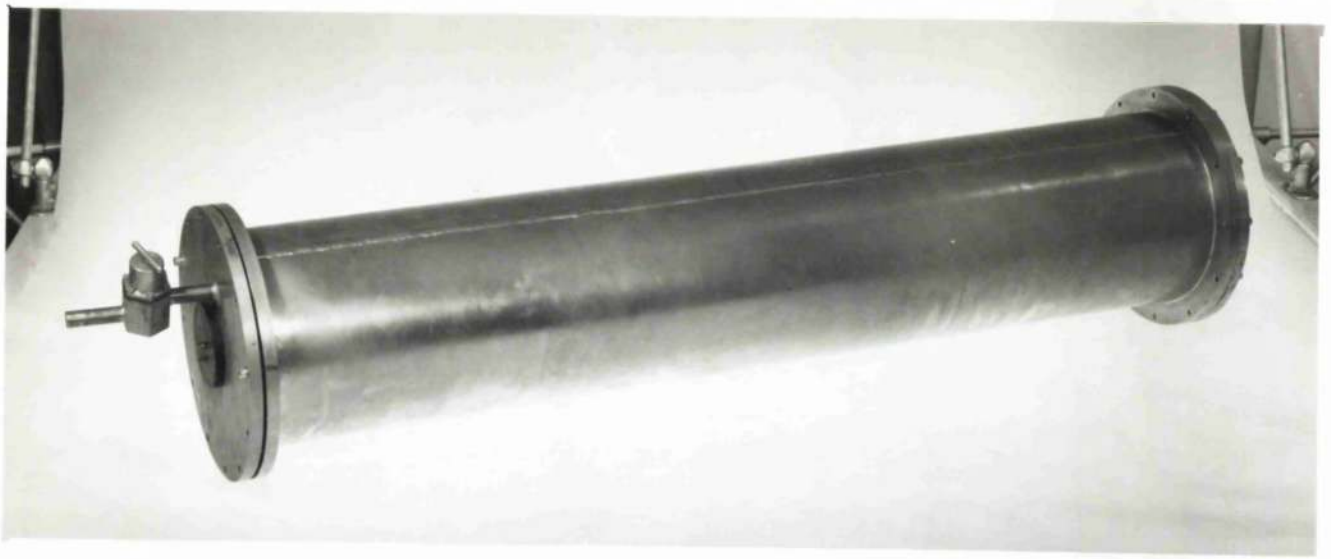


FIG. 4

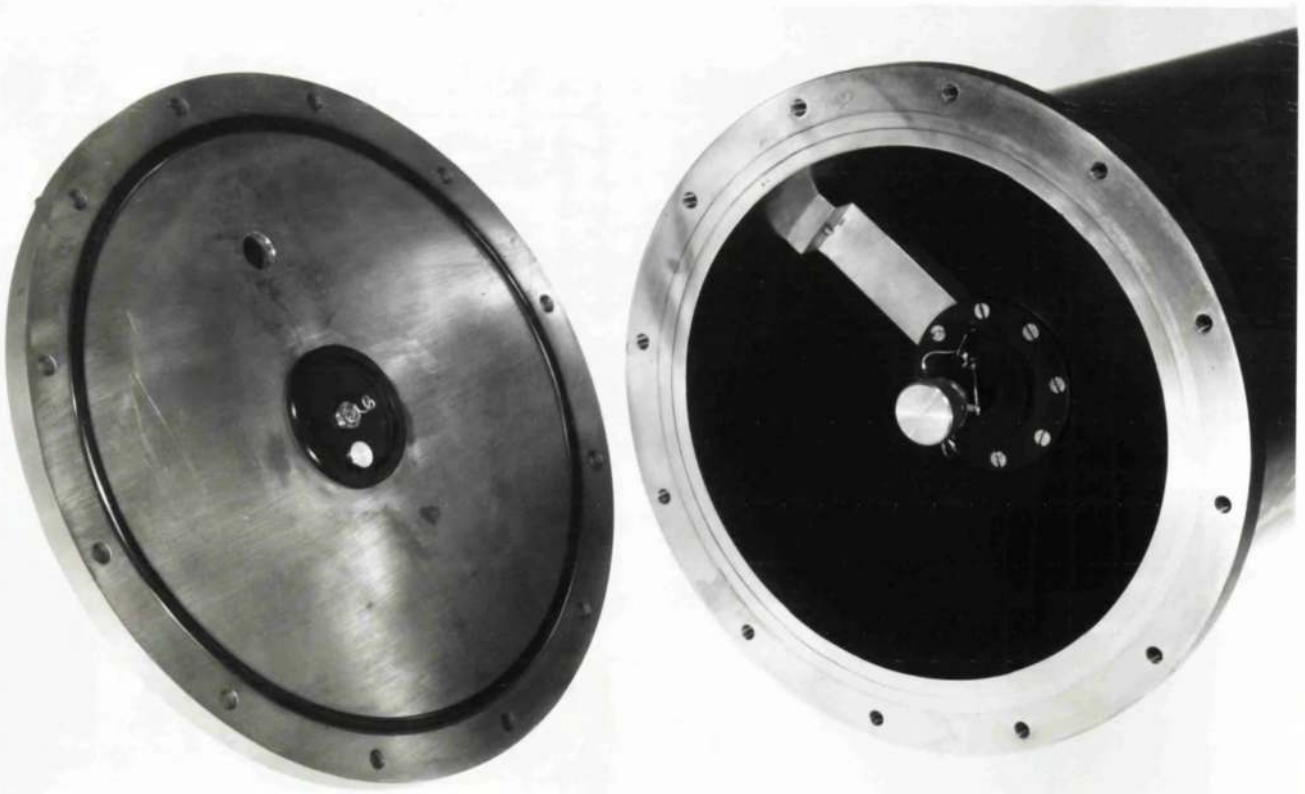


FIG 5.

The source is spread on this liner. Fig. 4 is a photograph of the counter and Fig. 5 shows an end-view of the counter with the field tube support and electrode connections. Fig. 6 is a typical counter electrode assembly.

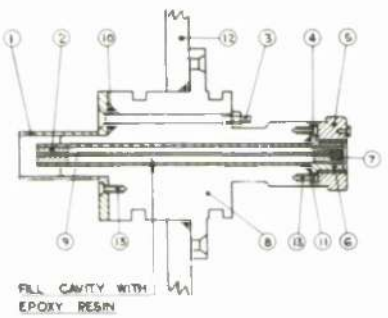
Calibration.

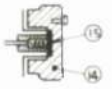
Since the length of the insensitive part of the counter is considerably longer than the range of the α-particles, it is found convenient to calibrate by having an electrodeposited source of mixed plutonium isotopes mounted on a piece of soft iron inside the counter. This can be moved in and out of the sensitive volume when desired by an externally placed magnet. For calibration of the longer-lived species it is intended to use a samarium oxide source chosen to be the same thickness as the unknown source. This avoids the effect of absorption on apparent energy discussed earlier.

The resolution of the Pu-236 peak was 5% full-width at half-maximum height when the instrument was filled to one atmosphere with argon plus 10% methane.

Results and Discussion.

0.935 gram of Sm_2O_3 was mounted on the copper cathode using the source mounting technique described for the ion chamber, care being taken that the source was completely within the sensitive volume of the counter. Preliminary examination of the spectrum revealed that the peak tended to drift with time. This was caused by the accumulation of charge on the source. Mixing the source with a small amount of colloidal/





① FIELD CORRECTION TUBE (BRASS.)
② INSULATING PLUG E.G. PERSPEX.
③ FIELD TUBE TERMINAL 8 B.A.
④ GUARD TUBE.
⑤ FINGER NUT AND GUARD TUBE TERMINAL.
⑥ INSULATING PLUG.
⑦ METAL INSERT (TO PERMIT ANCHORING OF ANODE WIRE BY SOLDERING.)
⑧ EBONITE HOUSING.
⑨ 0·020" HYPODERMIC TUBING.
⑩ O-RING B.S.1806 O.S.1.
⑪ O-RING B.S.1806 O.S.5.
⑫ COUNTER END FLANGE.
⑬ SCREW 8 B.A. x ¼" LONG.
⑭ INSERT SHOWING WIRE TENSIONING SPRING AT OPPOSITE END OF COUNTER.
⑮ NEOPRENE RUBBER GAS SEAL.

FIG. 6 PROPORTIONAL COUNTER ELECTRODE ASSEMBLY

FIG. 6.

/colloidal graphite proved an effective cure but unfortunately introduced additional contamination of the uranium-thorium series with the consequent increase in effective background. The latter involved a correction of 2% to the Sm-147 spectrum.

Assuming an abundance of 15.07% for the Sm-147 isotope and 2π geometry yields a half-life of $1.21 \pm 0.04 \times 10^{11}$ years, in satisfactory agreement with the ion chamber method.

Whilst it is apparent that the proportional counting method is suitable for this type of work provided precautions are taken against source charging effects, the level of α-emitting impurity in spectrographically pure materials is sufficiently large to be troublesome. The level has been measured to be less than one part in ten million for typical materials. (See Chapter 3 Section 3.3) When application of this method to the longer-lived isotopes is considered, then the tails of peaks caused by impurity activity could be sufficient to completely obscure lower energy activities. Photographic emulsion methods are better from this aspect since source self-absorption is very small and there is practically no low energy distribution to the peaks. However, evaporation of the source material onto large areas of the proportional counter cathode would give thin sources ($< 10 \mu$ gm) and still permit quantities of ~ 50 m.g.m. to be used. This would directly be competitive with nuclear emulsion techniques.

An attempt was made to detect the alpha rediation from Nd-144 using the techniques for the samarium investigation but only an upper limit of $\geq 5 \times 10^{15}$ years could be obtained because of the impurity level. This result is in disagreement with that of Waldron et al[25] and is bordering on the half-life quoted by Porschen and Reizler.

Senftle[32] et al have failed to observe α-activity from Ce-142. The lower limit of 10^{16} years is in disagreement with Reizler and Kauw[28]. On consulting Table II it is apparent that, with the exception of Sm-147 which is well known and the results on Pb-204 and Pt-190, there is either direct disagreement or the half-lives are unconfirmed. Further, Rasmussen[34] in his calculation for alpha decay barrier penetrabilities finds the results for platinum to be so anomalous as to cast doubt on the experimental data.

In view of the uncertainty in the data, and the danger of the natural uranium and thorium alpha activities being present, a complete re-investigation of these activities is merited.

TABLE I.

Predicted Disintegration Energy for β-stable Naturally Occurri Disinteitters among the medium-heavy Elements

Element Z	Mass Number	a Energy MeV	Element Z	Mass Number	a Energy MeV
erium 58	142	1.602	Tantalum 73	181	1.341
eodymium 60	144	2.367	Tungsten m74	180	2.232
	146	1.304		182	1.806
	148	0.442		183	1.592
				184	1.380
amarium 62	147	2.385		186	0.950
	148	1.953			
	149	1.523	Rhenium 75	185	1.843
	150	1.092			
	152	0.233			
			Osmium 76	184	2.735
uropium 63	151	1.419		187	2.093
				188	1.879
				189	1.665
adolinium 64	152	1.745		190	1.452
	154	0.883		192	1.042
	156	0.020			
			Iridium 77	191	1.914
ysprosium 66	156	0.655			
	158	0.082			
			Platinum 78	190	2.806
rbium 68	164	1.588		192	2.379
	166	1.161		194	1.950
	167	0.945		195	1.737
	168	0.733		196	1.522
	170	0.305		198	1.095
hulium 69	169	1.198	Gold 79	197	1.988
tterbium 70	168	2.089	Mercury 80	196	2.879
	170	1.661		198	2.452
	171	1.448		199	2.237
	172	1.233		200	2.024
	173	1.019		201	1.811
	174	0.806		202	1.596
	176	0.377		204	1.167
utecium 71	175	1.267	Thallium 81	203	2.062
afnium 72	174	2.162	Lead 82	204	2.528
	176	1.734		206	2.098
	177	1.520		207	1.882
	178	1.306		208	1.670
	180	0.877			

TABLE II

Summary of Experimental Results.

Method	E_α MeV	Years	Reference
		Ce - 142	
1. Nuclear Emulsion	1.5	5.1×10^{15}	W. Riezler and G. Kauw (28)
2. Nuclear Emulsion	-	$\geq 10^{16}$	F.E. Senftle, T.W. Stern V.P. Alenka (32).
		Nd - 144	
1. Nuclear Emulsion	1.9	2×10^{15}	E.C. Waldron, V.A. Schultz and T.P. Kohman (25)
2. Nuclear Emulsion	1.8	5×10^{15}	W. Porschen and W. Riezler (26,27).
		Sm - 147	
1. Nuclear Emulsion	2.12 ± 0.03	-	D. Szteinsznaider (18)
2. Nuclear Emulsion	2.29	-	G. Hinschberger, A. Starling and G. Ambrosino (19).
3. Nuclear Emulsion	2.18 ± 0.14	$1.15 \pm 0.03 \times 10^{11}$	G.E. Leslie (20)
4. Gridded Ion Chamber	2.19 ± 0.01	-	A.A. Vorober, A. Kosner, V.A. Korolev, G.E. Solyakin (21).
5. Calibrated Gridded Ion Chamber	2.20 ± 0.03	$1.14 \pm 0.05 \times 10^{11}$	M. Karras, M. Nurmia (22).
6. 4 proportional counter	-	$1.25 \pm 0.06 \times 10^{11}$	G. Beard and M.L. Wiedenback (23) See also E. Picciotto (24).
		Gd - 152	
1. Nuclear Emulsion	1.7	9.6×10^{14}	W. Riezler and G. Kauw (30)
		Dy - 156	
1. Nuclear Emulsion	-	$> 10^{18}$	W. Riezler and G. Kauw (29)
		Hf - 174	
1. Nuclear Emulsion	2.5	4.4×10^{15}	W. Riezler and G. Kauw (30-

TABLE II (Continued).

Method	E_α MeV	Years	Reference
		Tungsten ≥ 180	
$CdWo_4$ and $CaWo_4$ scintillation counting	-	$> 8K \times 10^{17}$ where K = rel abundance of the isotope.	G.B. Beard and W.H. Kelly (33)
Nuclear Emulsion	3.0	$= 2.2\ K \times 10^{17}$ years	W. Porschen and W. Riezler (27).
		Pt - 190	
Nuclear Emulsion	3.3	5.9×10^{11}	Th. Mayer - Kuckuk (31).
Nuclear Emulsion	3.3	9.6×10^{11}	W. Porschen, W. Riezler (26,27).
		Pt - 192	
Nuclear Emulsion	2.6	$\sim 10^{15}$	W. Porschen, W. Riezler (27).
		Pb - 204	
Nuclear Emulsion	-	$> 7 \times 10^{16}$	T.P. Kohman (17).
Nuclear Emulsion	2.6	1.4×10^{17}	W. Riezler and G. Kauw (29)

Chapter 5.

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Chapter 6.

Future Work.

It is apparent that in all cases, without exception, serious limitations on the sensitivity of the experimental arrangements are caused by the level of natural uranium, thorium and actinium impurities in the source material. In future work, therefore particular attention should be given to removing the U-Th-Ac group and their active decay products. (See Chapter 3 'The Half-Life of V-50' γ-spectra.)

Referring to the table of triple and double isobars in Chapter 1 and considering the results described in this thesis we see that the theoretical predictions regarding stability have been fulfilled with the exception of the Cd-In-113 pair and Ta-180. Calculation shows that Cd-113, if the active member of the pair, should have a half-life for β-emission $>5 \times 10^{15}$ years i.e. above the sensitivity of the present equipment. An order of magnitude improvement in the impurity level will almost certainly permit observation of the decay. Similar purity levels would be required for the 'blank' element used as a background. An alternative would be to obtain gram quantities of electro magnetically enriched isotopes. This would ensure a very high degree of purity but such large quantities are not available at present although may be in the future. A factor of ten increase in sensitivity would be attainable if this were the case. On the other hand, if In-113 were the active isotope then a half-life $>10^{17}$ years can be/

/be expected which is outwith the bounds of detection with present equipment. In addition separated isotopes would be required since In-115 has a relatively large β-activity which would obscure any x-ray spectrum. Corrections would also be required for the x-radiation induced by absorption of the β-rays in the source. If a gaseous form of indium can be found then use of the multi-wire counter[1] would greatly enhance the possibility of detection for a) the background would be extremely low and b) generation of x-rays by bombardment either with α, β radiation from impurities or the In-115 β radiation or by cosmic radiation would be removed by the internal anti-coincidence device or the 4π geometry would integrate the spectrum and carry any coincidence x-rays to higher energies. Such a system would enable half-lives of 10^{18} years to be measured provided the selected gas source is non-electron attaching. This latter requirement may prove the main difficulty.

Ta-180 was not examined in this series of experiments as its existance was only discovered during the work. It is intended to search for γ-activity with the large 9" NaI (Tl) γ-ray spectrometer in the near future. An order of magnitude at least, should be attainable over the 10^{13} year upper limit already reported[12].

The electron-capture, β-branching ratio for La-138 is worth re-measuring as results are not in sufficiently good agreement. With the development of large crystals in the past few years spectra free from Compton electron distribution/

/distribution would be obtained. This would simplify calibration and permit large source areas to give a high count rate without self-absorption corrections. The availability of beryllium - windowed NaI crystals would permit easy x-ray - γ-ray coincidence experiments to find the K electron capture half-life which should correspond to that obtained with the direct γ-measurement. Further work is required on the β branch which is of low intensity because of source absorption. An evaporated source over a large area proportional counter may help although correction for x-ray detection would be required if measuring the specific β activity. An accurate end-point energy should be obtained by this method.

The decay scheme of Lu-176 is not yet certain as the half-lives measured by β-emission and by the γ-rays are not in good agreement. There is some doubt about the existence of an electron-capture branch which is worth further investigation to obtain a branching ratio.

Application of the large crystal γ-ray spectrometer to the half-life measurement of V-50 provides a very sensitive technique. Re-examination of the spectrum using a much purer source should yield a conclusive answer and permit an actual determination of the γ-ray energies which have so far been implied from neighbouring decay schemes. The experiment described in Chapter 3 was also handicapped slightly by the presence of Co-60 impurity.

A very recent publication on the half-life and maximum energy of In-115 has raised some doubt about the latter value. The work described in this thesis seemed conclusive in that the maximum β energy of 480 ± 20 Kev have a good fit with theory and agreed with experiments on the metastable state [4]. Beard and Kelly[3], however, have confirmed the half-life at $6.9 \pm 1.5 \times 10^{14}$ years and obtained $E_\beta = 0.625 \pm 0.07$ Mev which is in good agreement with Libby's value of 0.63 ± 0.03 Mev obtained by absorption methods and disagrees with the above results. The large error in the new result is caused by the difficulty in calibrating the liquid scintillators used. In view of this large discrepancy a further independent measurement of the energy is desirable.

Apparently conclusive results were found for the half-life of Te-123 by four different experiments. The large log ft value, however, would indicate a third forbidden transition where as a second forbidden transition is predicted. Here, again, source purity proved a handicap and a check experiment would be worth while.

Re-187 is unique because of the very low disintegration energy (1.2 Kev). Because of this, source absorption was very large and prevented an accurate half-life measurement. Rhenium can be obtained in the form of a volatile compound, hence an accurate half-life should be attainable using the source in vapour form. This isotope is especially interesting because of its application to geochronology.

The proportional counter method of detecting very low specific activity α-emitters is very promising. The difficulty of obtaining pure sources is once again the limiting factor and if lower impurity levels can be attained, the field is wide open for an investigation of the expected natural α-emitters listed in Chapter 5 Table I. Upper limits of 10^{20} years are envisaged.

Double β-decay.

An important application of low background coincidence counting techniques is the study of double beta decay. This is a special type of very long-lived radioactivity predicted by beta decay theory in which a nuclide decays by the emission of two time-coincident beta particles, thereby increasing in atomic number by two units. The probability of the phenomena is dependent on the square of the second order terms in the matrix element, the first order terms being vanishingly small so that single beta decay will not occur. Two types of double beta emission are permitted by the general nuclear-lepton Hamiltonian. In the first two neutrinos are emitted simultaneously with two electrons and in the second no neutrons are emitted.[5,6]

Before the discovery of parity non-conservation[7], double beta decay provided a possible experimental test to decide whether the neutrino is a symmetrical particle existing in only one form (i.e. a Majorana particle) or a Dirac particle which could occur in two forms either as a neutrino(ν) or as an/

/an anti-neutrino ($\bar{\nu}$) analogous to the electron and positron respectively. If the neutrino is a Dirac particle then two anti-neutrinos must be emitted along with the two electrons

$$\text{i.e. } 2n \rightarrow 2p + 2e + 2\bar{\nu} \quad \text{- - - - - - - (1)}$$

Majorana, on the other hand, suggests that there are only two states for a given momentum of the neutrino corresponding to the two possibilities of the spin. In this case the neutrino can be either emitted or absorbed in the beta decay process.

$$\text{i.e. } n_1 \rightarrow p_1 + e_1 + \nu$$
$$\nu + n_2 \rightarrow p_2 + e_2$$
$$\text{giving } 2n \rightarrow 2p + 2e \quad \text{- - - - - - - (2)}$$

It follows that in 1 there will be a continuous energy spectrum with a maximum energy = E_o i.e. the total maximum kinetic energy of the beta particles and from 2 the spectrum will have a sharp peak at E_o i.e. a constant.

The Majorana theory predicts a half-life in the region $10^{13} - 10^{18}$ years for the transition whereas the Dirac theory expects the half-life to be about 10^{24} years. A measure of the energy spectrum or the half-life would therefore give a clear-cut answer on the nature of the neutrino.

Many attempts were made to detect the phenomena using various methods e.g. geological, cloud chambers, scintillation counting and photographic emulsions. These are fully summarised by Primakoff and Rosen[8]. In most of the experiments only lower limits were placed on the half-life and in cases where double beta decay was claimed to have been observed have since/

/since been discounted except the work of Inghram and Reynolds[9] who made an isotopic analysis of xenon extracted from geologically old tellurium ores. Excess xenon abundances were found for most of the isotopes and attributed to (n,γ) reactions on the Te isotopes. However, there was a residual excess for Xe-130 which was attributed to double beta decay in Te-130. The half-life is quoted at 1.4×10^{21} years which comes about mid-way between the two theoretical predictions and should rule out the possibility of the neutrino being a Majorana particle.

However with the discovery of parity non-conservation, it was at first thought that the intermediate lepton would not be absorbed by the second neutron because it would have the wrong longitudinal polarization and neutrinoless double beta decay could not occur[10]. Janouch[11] modifies this view by pointing out that the interaction constants in the Hamiltonian may not be equal (if equal double beta decay would be impossible) and could differ by 10-20% as the experimental determination of these constants is obtained by measuring the longitudinal polarization of the electrons, or the circular polarization of the γ-rays emitted by the excited product nucleus, which is only accurate to this amount. Hence neutrinoless double beta decay may still be possible since the second order matrix element does not vanish. Calculations for the half-life of Ca-48, on this basis yield a result of $T_{\frac{1}{2}} \leq 2 \times 10^{19}$ years. It is also shown that since the possible occurrence of small admixtures of Scalar and Tensor interactions with the Vector and Axial-/

/Axial-Vector interactions has not been disproved entirely then neutrinoless double beta decay can occur. In this case Ca-48 would have $T_{\frac{1}{2}} \leq 2 \times 10^{22}$ years. The experimental evidence does not exclude these possibilities.

Primakoff and Rosen interpret the general experimental evidence as favouring two-neutrino double beta decay. If neutrinoless decay occurs then the principle of the conservation of lepton charge is violated and the neutrino is a Majorana particle. If two-neutrino decay occurs then parity non-conservation creates two possibilities 1) The total lepton charge is conserved and the neutrino is a Dirac particle, or 2) The total lepton charge is not conserved and the neutrino is a Majorana particle but the nucleon-lepton interaction must have a two-component 'neutrino' type coupling. The ambiguity between these two cases makes it impossible to decide whether ν is a Majorana or Dirac lepton and whether the total lepton 'charge' is or is not, conserved. Rosen[12] has followed this outcome by considering the relation between the conservation of lepton 'charge' and the shape of the spectrum. He describes conditions under which certain values of the coupling constants will yield roughly equal probabilities for the occurrence of no-neutrino and two-neutrino double beta decay and in which the electron-coincidence spectrum will have a broad peak at $\frac{1}{2} E_0$ with a sharp peak at E_0, also. Under these conditions i.e. that parity non-conservation is non-maximal, careful study of the spectrum offers a test for lepton 'charge' conservation.

It is apparent, therefore, that in future work in double beta decay any experiment must be capable of measuring half-lives of the order 10^{20} years, i.e. a factor of at least 100 improvement on present techniques, and it is extremely desirable that the spectrum should also be observed. At present, only geological methods could hope to detect such long half-lives but have the disadvantage of not revealing the energy spectrum.

To provide the data in a reasonable time period (<1 year) counting methods would required a source of several grams of enriched material, which could be mounted in a manner preventing serious source absorption. Coincidence techniques would be necessary to keep the background at a minimum. Such an experiment appears impracticable at the present time.

Chapter 6.

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ABSTRACT OF THESIS

"THE RADIOACTIVITY OF NATURALLY OCCURRING ISOTOPES WITH Z < 82"

SUBMITTED FOR THE DEGREE OF Ph.D. BY

D.E. WATT

This thesis describes the techniques used and the results obtained in a search for the long-lived naturally occurring radioactive isotopes with atomic number < 82 and excludes the uranium, thorium and actinium series.

Consideration of nuclear energies and stability characteristics lead to the conclusion that in cases where three neighbouring isobars, or a double isobar, occur in nature then the odd-odd member of the triple isobar and one member of the double isobar must be radioactive. Since these nuclides are expected to have very long half-lives, greater than the age of the earth, highly sensitive detection techniques, capable of measuring low specific activities are necessary.

The nature of the cosmic radiation is considered in a fundamental approach to the form of shielding required to produce the optimum conditions for low background counting. Contributing factors, of a secondary nature, to the background counting rate in proportional and scintillation counters are discussed and suggestions are given on how these can be removed. A background of 0.33 counts/min/litre, above 500 eV, at one atmosphere pressure, was obtained for a wall-less proportional counter /

counter with an arrangement using boron-loaded paraffin wax inside a low background laboratory. It is concluded that this residual count rate is due to γ-radiation and that it would be greatly reduced with a more efficient γ-ray detector in the anti-coincidence system.

The characteristics of an underground laboratory and of a specially built low-background cell are described.

With the above experimental arrangements a search was made for natural radioactive species with atomic numbers below 82 and detailed investigations were made of the triple isobars (La-138; V-50; and Lu-176) and of the double isobars (Cd-In-113; In-Sn-115; Sb-Te-123; and Re-Os-187).

The existence of two γ-rays at 0.81 ± 0.01 MeV and 1.44 ± 0.02 MeV in La-138 was confirmed. No annihilation quanta were observed. Coincidence studies show that these γ-rays are not in cascade but that the 1.44 MeV transition is in coincidence with the electron capture branch. A previously unobserved β-ray with maximum energy of 205 ± 10 KeV was detected. The partial half-life of the electron capture branch is found to be $(1.64 \pm 0.06) \times 10^{11}$ yea From the β-ray specific activity the half-life is $(4.1 \pm 0.7) \times 10^{11}$ years and from the 0.81 MeV γ-ray is $(3.5 \pm 0.3) \times 10^{11}$ years. A log ft value of 19 is calculated for the β branch indicating that the transition is 3rd forbidden.

Two experiments were carried out on V-50. In the first a half-life of $(4.0 \pm 1.1) \times 10^{14}$ years was obtained using a two inch NaI crystal to detect the γ-rays from a sample known to contain the normal amount of the Uranium-Thorium-Actinium series, as impurity. Later a re-investigation of this isotope using a combination of a 9 inch x 6 inch NaI crystal with five kilograms of spectrographically pure materials, which gave a factor of twenty increase in sensitivity over the best of the previous methods, proved the half-life to be $1.8 \pm 0.4 \times 10^{16}$ years for negatron emission and $9.6 \pm 1.6 \times 10^{15}$ years for electron capture. It is shown that the degree to which natural uranium and thorium occur in the source material must be accounted for in the measurement of very low specific activities.

The importance of correcting for changes in the natural background spectrum shape caused by the different absorption for the cosmic radiation and local radioactivity in the presence of large amounts of source material is emphasised.

A search was also undertaken for γ-rays or conversion electrons which may be associated with the expected, but so far unobserved, electron capture branch of Lu-176. It is concluded that the upper limit for this mode of decay is $(3 \pm 1)\%$. The half-life of the branch obtained via

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measurements with the associated γ-rays is $(2.1 \pm 0.2) \times 10^{10}$ years.

Among the double isobars investigations on Cd-113 and Sb-123 show these to have minimum β half-lives of 1.3×10^{15} years and 1.32×10^{16} years respectively. A new value of 480 ± 30 KeV has been obtained for the beta disintegration energy of In-115 and the half-life was confirmed to be $6.0 \pm 0.4 \times 10^{14}$ years. The K electron capture of Te-123 was observed for the first time. A half-life of $1.24 \pm 0.10 \times 10^{13}$ years is calculated. The beta decay of Re-187 was found to have a maximum β energy of 1.2 ± 0.10 KeV and an estimated half-life of 3×10^{10} years. This is the softest β-ray spectrum ever to be detected. Lower limits for gamma ray emission from In-113, Sb-123 and Te-123 are respectively 1.4×10^{15} years, 1.5×10^{15} years and 2.5×10^{13} years.

The systematics of natural α-emitters among the medium heavy elements are discussed and a proportional counter method with the potential of measuring α half-lives up to 10^{20} years is described. Limitations on this method are the levels of natural uranium, thorium and actinium impurities, in most substances.

Finally, suggestions are given for some future experiments which should clarify the position regarding

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the natural radioactive substances below lead. The significance and possible detection of double β decay is considered.